PETROLOGY AND EVOLUTION OF THE NORTHERN PART OF THE RHUM ULTRABASIC COMPLEX

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The Rhum Cuillin - Barkeval,Hallival,Askival,Trollval

Not a seat, but a springboard.

Winston L. S. Churchill, 29th November, 1942.

ABSTRACT

The central region of the Rhum ultrabasic complex comprises three major sequences of olivine, and plagioclase-olivine cumulates: (i) the westerly continuation of the Eastern Layered Series; (ii) the northernmost extension of a redefined Western Layered Series, and (iii) a newly-defined Central Series. The structural relationship of the Eastern and Western Layered Series is unresolved, but they are transgressed by the large dyke-like mass of the Central Series. The Long Loch Fault played a significant role in the development of the essentially in situ Central Series. Striking differences between the three series reflect fundamental differences in the way they accumulated and solidified.

Field, textural and geochemical evidence favour a multiple intrusion origin for the macro-layering. Gravitational deposition from flowing magma currents, and in situ crystallization, produced the fine-scale layering of the plagioclase-olivine and olivine cumulates respectively. A complex pattern of cryptic variation in both the cumulus and intercumulus phases supports the magma replenishment model.

Field, petrographic, geochemical and experimental studies of the transitional basalt → picrite dyke suite have demonstrated the existence, on Rhum, of high-magnesian liquids with up to 20.5% MgO. The suite as a whole is characterized by low incompatible-element contents and slightly LREE-enriched chondrite-normalized REE patterns. The high-MgO liquids are believed to be the result of small degrees of melting (15-20%) of garnet lherzolite at a depth of ca. 65 km, leaving a lherzolite residuum. Similar magnesian liquids were probably parental to the ultrabasic complex.

The nature of the magma entering the Rhum chamber varied greatly. Magma supplying the chamber during the development of the Eastern and

Western Layered Series carried only minor amounts of liquidus olivine (and spinel) - ca. 10%. In contrast that supplying the Central Series chamber was highly olivine-phyric (ca. 50% crystals). Intrusive offshoots from the Central Series porphyritic (picritic) magma are represented by the numerous non-layered peridotitic minor intrusions, and low-temperature (basaltic) magma by the gabbroic minor intrusions.

Available evidence suggests that mantle fusion in the Rhum area pre-dated that of Skye. Close chemical resemblances between the picritic - basaltic magmas of Rhum and the magnesian basalts from known Tertiary spreading centres suggest there were genuine, but abortive, attempts to produce oceanic crust beneath Rhum.

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CHAPTER 1

INTRODUCTION

1.1 Regional Setting

The Isle of Rhum is one of a series of deeply-eroded Tertiary subvolcanic igneous complexes along the west coast of Scotland (fig 1.1). It is the largest and most spectacular of the group of islands forming the Parish of the Small Isles, Inverness-shire; its striking mountainous skyline contrasting with the flat low-lying profiles of neighbouring Eigg, Muck and Canna. Roughly diamond-shaped in plan, and measuring 13 km N-S and 12 km E-W, Rhum is situated 15 km south of Skye and 20 km NW of Ardnamurchan.

Ten major igneous centres compose the British Tertiary Igneous Province (fig 1.2); the Province is comparable in age and very general tectonic significance to (a) Baffin and W Greenland, (b) E Greenland (S of 70°N) and Faeroes, (c) NE Greenland, and (d) Skaggerak.

1.2 Local Setting

Marine geophysical surveys and limited sampling in the Sea of the Hebrides have shown that the Isle of Rhum is situated in an asymmetrical sedimentary trough (bounded by the Minch and Camasunary-Skerryvore Faults) which contains approximately 2.5 km of Mesozoic strata underlain by an unknown thickness of westward-dipping Palaeozoic and Precambrian sedimentary rocks (Binns *et al.*, 1974). The Triassic outlier in NW Rhum probably represents the eastern edge of this Minch Mesozoic basin (see fig 1.3). The Palaeocene-Eocene lavas of Skye extend offshore into the axial region of the trough, and NW of Canna they contain a basin of probable Oligocene-age sediments (Smythe and Kenolty, 1975).

Bathymetric studies show the Sea of the Hebrides to consist of a complexly-glaciated shelf dissected by overdeepened hollows to form a series of ridges and basins. A strong NE trend is evident (Binns *et al.*, 1974).

The Bouguer anomaly gravity map of Rhum produced by McQuillan and Tuson (1963) shows a large positive anomaly of +76.3 milligals centred over the ultrabasic complex. Assuming densities of between 3.05 and 3.10 gm/cm^3 for the basic and ultrabasic rocks, and a local crustal density of 2.75 gm/cm^3, the gravity data suggest two alternative models: a) a vertical cylinder of 10 km diameter extending 15 km downwards; b) a truncated cone with a surface diameter of 8 km, extending down to 15 km, with sides inclined outwards at 77° to the horizontal. Model (b) is preferred because it fits the observed size of the ultrabasic complex. The gravity data show the intrusion to have no important horizontal extension beyond the limits indicated by the exposed rocks, and a substantial portion of the complex must lie under the sea to the south of the island.

1.3 Geology of Rhum

Topographically and geologically Rhum can be divided into two parts. The low-lying northern half and eastern coastal strip consist of a thick sedimentary succession of Proterozoic arenites (Torridonian Formation), whereas the southern more mountainous part of the island comprises a Tertiary intrusive central complex emplaced along an arcuate ring fracture - the Main Ring Fault. A simplified geological map of Rhum is shown in fig 1.4. Comprehensive summaries of the geology have been given by Stewart (1965), Dunham and Emeleus (1967) and Emeleus and Forster (1979) (see Table 1.1). Consequently only a very brief resume of the geology is given here.

1.3.1 Pre-Tertiary

The oldest rocks on Rhum are Archean (Lewisian) gneisses. These interbanded leucocratic and mafic gneisses (and subordinate amphibolitized metabasite dykes and sheets) outcrop at several localities within the area bounded by the Main Ring Fault (Bailey, 1945). The gneisses frequently show signs of extensive thermal metamorphism, and at two localities they are unconformably overlain by basal Torridonian sediments.

Approximately 4.4 km of Torridonian grits, shales, and arkoses occur as a westerly dipping sequence in northern Rhum. The lowest division, the Basal Grit, occurs only within the Main Ring Fault, has been interpreted as a piedmont fan deposit (Black and Welsh, 1961), and is everywhere in faulted contact with the shales of the younger Bagh-na-h Uamha Division. These shales comprise thinly plane- and cross-laminated siltstones (possible floodplain deposits) and are conformably overlain by very thick trough-crossbedded arkoses (braided river deposits). The Rhum Torridonian succession has been correlated with that of Skye and the Mainland (Black and Welsh, 1961; Stewart, 1965).

Triassic sandstones, fossiliferous shales, and breccias form a small outlier in NW Rhum. Basal cornstones mark the unconformity with the Torridonian arkoses (Bailey, 1945).

Jurassic rocks on Rhum are restricted to an isolated fault-slice along the Main Ring Fault (Dunham and Emeleus, 1967) although fossiliferous Jurassic sediments occur on the neighbouring islands of Eigg, Muck and Skye. Upper Cretaceous sandstones resting unconformably on the Jurassic sediments on Eigg (Harker, 1908a), are absent from Rhum.

1.3.2 Tertiary

Tertiary rocks on Rhum are almost entirely igneous in origin, comprising lavas, ultrabasic, basic and acid major and minor intrusives, and agglomerates (Table 1.1). Tertiary sediments (river conglomerates and sandstones) intercalated with the lavas will be discussed more fully in Chapter 6.

The earliest activity appears to have involved intrusion of gabbroic plugs, now seen as partly shattered outcrops in the northern part of the complex (Emeleus and Forster, 1979). These early intrusions, together with their country rocks, were subsequently brecciated by the degassing of subjacent acid magma, to form explosion breccias and tuffisites (Hughes, 1960b; Dunham, 1968).

Directly following this explosive phase of activity, part of the acid magma was intruded as plugs and sheets of flow-banded porphyritic felsite. The remainder cooled slowly to form the large Western Granophyre and several smaller granophyre bodies; these granophyres were subsequently faulted up to the present level by the Main Ring Fault. A small exposure of Lewisian gneiss caps the Western Granophyre on Ard Nev and is thought to represent part of the original roof to the granophyre (Dunham and Emeleus, 1967).

Intrusion of the granophyres was followed by approximately 3 km uplift of the early acid rocks and their country rocks within the Main Ring Fault (Bailey, 1945). This major tectonic feature (diameter approximately 9 km), previously interpreted as a Palaeozoic thrust (Harker, 1903 and 1908a), is used to define the Rhum Igneous Complex. Outside the fault the Torridonian sediments show signs of disturbance, in the form of a swing in the regional strike, a steepening of sedimentary dips, and folding.

The highest peaks on Rhum (Barkeval, 591 m; Hallival, 722 m; Askival, 812 m; Trollval, 533 m; and Ruinsival, 521 m) were carved by Pleistocene glaciation from a large stratiform ultrabasic intrusion: a total of nineteen major rhythmic units are revealed, although the base is not seen and the roof has been lost through erosion. These units were labelled 1 to 15 (Brown, 1956) and A to D (Wadsworth, 1961). It is generally accepted that each unit formed by the gravitational settling and/or growth of spinel, olivine and plagioclase crystals on the floor of a repeatedly emptied and replenished subcrustal magma chamber. Marginal relationships suggest the layered ultrabasic rocks crystallized and consolidated at a lower level than they occupy at present, and were emplaced in an essentially solid condition along a ring fracture (the Inner Ring Fault) which was in places coincident with, and elsewhere transgressive to, the earlier Main Ring Fault. Basaltic magma which, according to Brown (1956) lubricated the ultrabasic mass during emplacement, solidified to form the Marginal gabbro. This forms an outer sheath around much of the layered complex, up to 200 m wide. Wadsworth (1961) has shown that the emplacement was later than the formation of the Western Granophyre: acid material back-veins the ultrabasic rocks, and basic cone-sheets in the granophyre are truncated, hornfelsed and sheared. The amount of uplift of the ultrabasic complex is indeterminate. From the lack of tectonic disturbance (layering in the ultrabasic rocks is well developed <1 m away from the granophyre) the uplift may have been quite small. Metamorphism of Torridonian arkoses, to produce tridymite, (since inverted), is shown to have taken place at ca. 3 km depth maximum (Brown, 1963). This figure would be equivalent to the depth of cover under which the uplifted ultrabasic complex was emplaced, and

hence if the assumption of a small uplift is correct, the original top of the magma chamber may not have been much deeper.

Intrusion breccias are a spectacular feature of the Tertiary geology of Rhum, and are found wherever the Marginal gabbro is in contact with felsic country rocks. At these contacts, breccia zones up to several metres in width, consist of blocks of mafic rocks in a fine- to medium-grained acid matrix derived from the adjoining granophyre, felsite, leucocratic gneisses or arkosic sandstones. The mafic blocks consist predominantly of disintegrated basaltic minor intrusions (individual blocks may still retain their chilled edges), though subordinate blocks of Marginal gabbro and peridotite also occur. These breccias provide evidence that the later mafic magma was able to produce rheomorphic acid magma from the country rocks, if only on a limited local scale (Dunham, 1964).

Throughout the volcanic history of Rhum, minor intrusions were being emplaced (Emeleus and Forster, 1979; Forster, 1980). Predominantly basic to ultrabasic, the majority occur as dykes and inclined sheets. The earliest appear to have pre-dated the tuffisites and explosion breccias, but many were intruded before and during the formation of the ultrabasic subvolcanic complex. A few cut the lavas and are the youngest intrusive rocks on Rhum. Acid minor intrusions are rare and appear closely related in space (and time?) to the granophyres. In addition to the dykes and inclined sheets, numerous coarse-grained plugs of gabbro and peridotite intrude the Torridonian rocks in northern Rhum, on both sides of the Main Ring Fault, and also cut the ultrabasic layered series (Table 1.1).

Tertiary lavas and interbedded sediments form small outliers on the hills of NW Rhum (fig 1.4). The lavas form four chemically and

temporally distinct groups erupted in the sequence alkali basalt → hawaiite → basaltic andesite → icelandite (Emeleus, 1976). Direct observations, and the contents of the interbedded sediments, indicate that the lavas post-date a) the emplacement, and b) the exposure of the granophyres and ultrabasic rocks (in direct contrast to other centres in the British Tertiary Province, where the lava piles pre-date the central complexes, e.g. Skye, Mull). The lavas overlap the Main Ring Fault south of Fionchra, lie directly on a granophyre surface on the north side of Orval, and on Fionchra and Bloodstone Hill can be seen to infill valleys carved from Torridonian sediments, the Western Granophyre and, sometimes, from earlier flows (Black, 1952b; Emeleus and Forster, 1979). Their unmetamorphosed condition relatively close to the ultrabasic mass is also suggestive of their young age.

The coarse intra-lava conglomerates contain boulders and pebbles of Torridonian sandstone, gneiss, granophyre, felsite, explosion breccia, tuffisite, gabbro and basalt, together with highly feldspathic ultrabasic rocks (Dunham and Emeleus, 1967) and scarce rotted peridotites (Upton, pers. comm.) which are interpreted as derivatives of the layered ultrabasic complex. Because the majority of rock types found in the Central Complex are represented amongst the conglomerates, the complex must have been unroofed and quite deeply dissected during Tertiary times, undergoing active erosion during the accumulation of the lavas (as much as 2-3 km of cover was eroded from the complex during this time). The size and well-rounded nature of the clasts indicates a very high energy environment suggestive of deep gorges, occupied by fast-flowing rivers bringing detritus from the high ground.

The source of the lavas is not known. They cannot be linked with the older lavas of Eigg and Muck (cut by a pre-ultrabasic complex dyke

swarm). However the occurrence of granophyric pebbles, similar to those on Rhum, in the conglomeratic beds within the Canna volcanic succession, is suggestive of contemporaneous lava extrusion on Canna and Rhum (Emeleus, 1973). Granophyre pebbles are also reported from the conglomerates near the base of the Skye Main Lava Series. These pebbles cannot be derived from the Skye granophyres (the Redhills Complex post-date the lavas (Bell, 1976)) but their petrographic similarity to the granophyre pebbles from the Rhum and Canna conglomerates (Williamson, pers. comm.), and the subsea connection between the Canna and Skye lava successions (Binns *et al.*, 1974), suggest they are derived from Rhum. If so, the Rhum lavas were probably extruded contemporaneously with the lowermost lavas exposed on Skye.

1.3.3 Comparison with other Hebridean Tertiary igneous centres.

From amongst the range of British Tertiary central igneous complexes, the Isle of Rhum is of special interest for several reasons:

(a) there is no direct evidence that a central volcano existed in Rhum: in situ extrusives from an early basaltic phase are absent (?lost through erosion). The local igneous history began with early intrusion of small gabbroic plugs, followed by explosive and intrusive acid magmatism. Consequently the major intrusion was emplaced into Precambrian country rocks, and the representatives of this early acid phase - not into the base of a volcanic pile, as in Skye and Mull.

(b) the relative proportions of rock-types within the Rhum central complex differ significantly from other centres: the large layered ultrabasic-basic intrusion predominates. In contrast the ultrabasic rocks within the Skye Cuillin complex are subordinate to layered gabbros (Bell, 1976); in Arran, Mourne Mountains and Slieve Gullion complexes granites and granophyres are dominant (Hatch *et al.*, 1974).

Even the Blackstones complex, with its large positive gravity anomaly (+ 135 mgals), does not show abundance of ultrabasic rocks in outcrop (Durant *et al.*, 1976).

(c) Shift of the focus of activity, a characteristic feature of many British Tertiary complexes, is not apparent in Rhum: detailed mapping has not established the existence of several multicomponent major centres of intrusive activity, as found in e.g. Skye, Mull and Ardnamurchan (Richey, 1961; Bell, 1976).

(d) Rhum does not show the extensive development of ring-dyke and cone-sheet intrusions (cf. Mull, Ardnamurchan and Skye).

(e) Rhum is probably amongst the earliest of the Tertiary U.K. centres: if the granophyric pebbles from the conglomeratic beds within the Skye volcanic succession are derived from Rhum, the Skye complex must be considerably younger than Rhum (see section 1.3.2 and Chapter 6).

Rhum's ultrabasic-basic complex is unique within the British Tertiary Volcanic Province because of:

(i) the dominance of ultrabasic over basic rocks;

(ii) the conspicuous, well-developed macro-rhythmic layering in the eastern half of the complex, involving alternating olivine- and plagioclase-olivine cumulates (Chapter 2);

(iii) widespread development (especially in western and central portions) of skeletal and dendritic (disequilibrium) olivine and plagioclase growth patterns ("harristic" olivine and "poikilo-macrospherulitic" feldspar textures - see Chapter 2);

(iv) extensive occurrence of ultrabasic breccias (Chapter 2).

1.4 Previous Research

The relatively small Isle of Rhum has been a focal point of geological attention since the early nineteenth century. MacCulloch made the first observations in 1819 and 1824, and Judd's (1889) contributions included the concept that Rhum represented the eroded root of a central-type volcano. Harker was the first to systematically map the island, and his work culminated in the Geological Survey Memoir on the Small Isles (1908a) and the accompanying one-inch Geological Map (1908b). For the next thirty-five years geological work on the island was limited, partly due to difficulty of access; however Phillips (1938) studied mineral orientations in the peridotites, and Tomkeieff (1945a) described the lava succession. In 1945 Bailey stimulated further research on the island with his classic paper which featured several major reinterpretations of Harker's conclusions on the Tertiary igneous tectonics. The paper prompted Tilley (1944) to reinvestigate Harker's collection of gneisses, and Tomkeieff (1945b), the ultrabasic rocks. In contrast to Harker's suggestion that each unit within the layered ultrabasic sequence represented a separate injection of magma (previously differentiated into olivine-rich and felspar-rich fractions), Tomkeieff (1945) suggested that there had been a single injection of heterogeneous magma, with streaking out of the components to form the layering. Wager's studies on Skaergaard led him to suspect that the celebrated layered rocks of Rhum could also be products of similar processes of differentiation of homogenous magma. Wager and Brown's ideas (1951) were developed by Brown (1956) who made a detailed study of the layered rocks in the Hallival-Askival area.

The lavas were again the subject of investigation, in the nineteen fifties, when Black published work on the volcanic succession

(1952a) and its age relations with the granophyre (1952b). Further petrological accounts have been given by Ridley (1971, 1973) and Emeleus (1976). In 1952(b) Black suggested that the Rhum granophyres originated through granitization of the Torridonian country rocks. However, the transitional contact between granophyre and Torridonian arkoses, claimed by Black, was proven to be faulted along its entire length (Hughes *et al.*, 1957).

Detailed revision of the Central Complex continued as Hughes remapped the Southern Mountains acid complex (1960b), Wadsworth remapped the layered ultrabasic sequence in SW Rhum (1961), and Dunham mapped the NE margin of the complex (1962, 1964, 1965, 1968). In the last 15 years there has been a wealth of published papers dealing with Rhum's ultrabasic breccias, olivine fabrics, rhythmic layering, cryptic variation, disequilibrium textures and possible parental magma compositions (Brothers, 1964; Dunham, 1965; Wager and Brown, 1968; Donaldson *et al.*, 1973; Donaldson, 1974, 1975a, 1975b, 1976, 1977; Gibb, 1976; Henderson and Gijbels, 1976; Maaloe, 1978; Williams, 1978; Dunham and Wadsworth, 1978; Forster, 1980; Huppert and Sparks, 1981).

Consequently the geological evolution of the Rhum Central Complex is known in sufficient detail to provide a fairly complete story. Several workers have compiled detailed summaries of the age relations and the petrogenetic and tectonic evolution of the Rhum complex (Dunham and Emeleus, 1967; Emeleus and Forster, 1979) - see Table 1.1.

1.5 Present Investigation

1.5.1 Introduction

In 1951 Wager and Brown produced a preliminary account of the rhythmic layering in the ultrabasic rocks. Brown (1956) continued this work with a detailed study of the Hallival-Askival sequence (the

Eastern Layered Series), noting that, further west, disturbances during the period of accumulation had produced slumping and brecciation of the layers. Attention then switched to the Harris area in the SW because of interest in the remarkable "harrisite" peridotite textures described by Harker (1908a). ("Harrisite" is a variety of peridotite characterized by very large and skeletal-shaped olivine crystals.) The rhythmic layers or "units" of this Western Layered Series are much thicker than those mapped by Brown; lack the highly felspathic cumulates so typical of the Hallival-Askival area; show a more marked development of "harristic" cumulates; contain zones of breccia absent from the Hallival-Askival region; and contain slightly more magnesian olivines near their bases. From these differences Wadsworth (1961) concluded that the Western Layered Series represented a distinctly lower level of the original intrusion than the sequence in the east, and that the relative displacement of the two series had occurred during the tectonic elevation of the ultrabasic complex. This displacement was believed to have taken place along the Long Loch Fault, a major topographic feature extending from Kilmory to Papadil (fig 1.4), which has an estimated easterly downthrow of approximately 1500 ft (Dunham and Emeleus, 1967).

However, none of the differences between the ultrabasic sequences east and west of the Long Loch Fault (listed above) were found in the Ruinsival region and further work was clearly required in Glen Harris, and on Trollval and Barkeval, to join up the Hallival-Askival and Harris Bay areas. As can be seen from fig 1.5 a large central portion of the complex, on either side of the Long Loch Fault, has remained uninvestigated since Harker's mapping for the 1908 Memoir.

1.5.2 Research objectives

The present research programme was initiated to help complete the mapping coverage of the ultrabasic rocks, begun in the late 1950's; it follows the work of D. Godfrey (prior to his tragic death in 1973). Further work to the south is now being undertaken by J. Volker (Edinburgh).

The objectives of this project are summarized below:

a) to produce a detailed and accurate geological map of the Barkeval - An Dornabac region (as outlined in fig 1.5);

b) to investigate the lateral extent of the Hallival-Askival layered sequence westwards across Barkeval, and the extent of the Western Layered Series north and eastwards across the Long Loch Fault, and hence correlate, if possible, the two halves of the complex;

c) to reconsider the origin of the ultrabasic rocks in the light of modern theories;

d) to formulate and present petrogenetic and tectonic models of the evolution of the Rhum Tertiary Layered Ultrabasic Complex, with particular reference to the physio-chemical processes occurring within magma chambers.

An initial two week reconnaissance survey in the autumn of 1977 was followed by a total of five months fieldwork during the summers of 1978 and 1979. This included the collection, by systematic vertical and lateral sampling, of a large representative suite of rocks for subsequent petrographical, mineralogical and geochemical studies at Edinburgh. Map 2 (back of Volume 2) shows the localities of all the samples collected.

Mapping was carried out on Meridian Airmaps Ltd. vertical aerial photographs having a scale of approximately 1:11000, and the details transferred to Ordnance Survey 1:10000 sheets. The geological map

has been drawn on a scale of 1:10000 - see Map 1 at back of Volume 2.

1.5.3 The study area

The position of the study area is shown on fig 1.5, and place names on Map 1. The western margin of the area has been taken as the contact between ultrabasic rocks and granophyre and Torridonian on the lower slopes of Ard Nev and Minishal respectively, and the eastern boundary is the centre of Coire Dubh south to Atlantic Corrie. To the south mapping has been extended to An Dornabac and Glen Harris. Thus defined, the project area includes all the ground between Coire Dubh, Minishal, An Dornabac and the floor of Atlantic Corrie, an area of approximately 16 km^2 and measuring at its maximum extent 4.5 km E-W and 4.5 km N-S.

Topographically the region can be divided into several subareas. The eastern half is dominated by Barkeval (591 m). Forming part of the Rhum Cuillin, Barkeval has an elongate summit plateau, and a southern face which drops steeply into Atlantic Corrie and is marked by a series of near vertical gullies, cliffs and active scree fans. Terrace features on the steep northern slopes are replaced on the western flanks by ridged and hummocky terrain.

The western half of the study area, from the western flanks of Barkeval to Loch an Dornabac, comprises, in the main, lower-lying ground (100-300 m above sea level) with a N-S trending ridge-and-hollow topography. A broad flat valley, the Stable Flats, occupies the NW corner of the area.

Evidence provided by striae and erratics indicates that the island was largely, if not entirely, overridden by W- and NW-moving ice sheets from the mainland during Pleistocene times (Peacock, 1976). A period of local glaciation subsequent to the decay of the last ice-sheet has long been recognized (Harker, 1908; Charlesworth, 1955;

Ryder, 1975) and recently Ballantyre and Wain-Hobson (1980) have found evidence for eleven former glaciers (fig 1.6). Coire Dubh and Atlantic Corrie are fine examples of glacial corries, floored by moraines and drained by extensive networks of streams. Those of Atlantic Corrie drain into the main Abhainn Sgathaig which flows south out of Long Loch (or Loch Sgathaig).

Exposure is generally good though complete exposure through vertical traverses is not often found, due to the easy weathering of the olivine-rich layers. Contacts between rock types are commonly obscured by vegetation.

CHAPTER 2

THE ULTRABASIC COMPLEX

2.1 General Introduction

In their comprehensive survey of layered intrusions, Wager and Brown (1968) recognized only three examples in which Fo-rich olivine-bytownite assemblages are dominant members of ultrabasic/ultramafic successions (all are from the British Tertiary Province - Rhum, Skye Cuillins, Ben Buie, Mull): Wilkinson *et al.* (1975) have since described similar bimineralic ultrabasic assemblages from the Salt Lick Creek intrusion, Western Australia.

The Rhum intrusion is an apparently unique representative (see Chapter 1, section 1.3.3) of a number of basic-ultrabasic intrusive centres in west Scotland (fig 1.1). The rocks are well-exposed with a roughly circular outcrop some 7 km in diameter. Olivine and plagioclase-olivine cumulates crop out along the eastern portion and dip gently (av. 20°) towards the centre of the complex. Western and central areas comprise predominantly olivine-rich ultramafites (similarly dipping towards the centre). Field evidence does not indicate the form of the original intrusion. It has been tectonically emplaced along a steeply-inclined arcuate ring-fracture (the Inner Ring Fault IRF, now marked by later intrusive gabbros - fig 2.1). A marginal facies is not developed, the base is unexposed, and the roof has been lost through erosion.

2.1.1 Cumulus theory : a review

The fractionation processes and crystallization conditions in deep-seated magma chambers cannot be observed directly, but the cumulates in stratiform intrusions retain a record (complete or partial) of these processes. Even after ca. 40 years problems still exist in explaining mechanisms that produced the effects of differentiation

seen in the field. Studies of layered intrusions frequently tend to underemphasize the fact that cumulates show only the very end effects of a long cooling history. Existing hypotheses are to a large extent ad hoc, and inadequately explain all observed features: the fact that many of the current theories have not changed greatly is testimony to the thoroughness of earlier workers.

The concept that basic magmas undergo fractional crystallization through crystal settling, first advocated by Darwin (1844) and theoretically demonstrated by Bowen (1915), gained popularity with the widespread recognition of igneous layering in several large plutons, e.g. Wager and Deer (1939). Convincing analogies drawn between structures in igneous and aqueous sediments (e.g. Wadsworth, 1973) strengthened the argument for the "cumulus theory".

The term "cumulus theory" refers to the concept that crystals nucleate (homogeneously) in one part of the chamber and are moved, gravitationally or otherwise, to another region. Opinions differ as to (a) the site and nature of the cumulus crystal nucleation and (b) the mechanisms of transportation to, (if necessary) and deposition at, their final resting place.

(a) Roof zones were regarded as sites of maximum heat loss, and therefore maximum crystallization (Hess, 1960; Wager, Brown and Wadsworth, 1960). In contrast, consideration of the positive pressure-dependence and relative values of adiabatic and liquidus temperature gradients (Jackson, 1961), and the uneven distribution of volatile components (Elsdon, 1970) implies maximum crystallization near the floor of the magma chamber. Additional consideration of possible convection paths (convection having a dominant role in heat and mass transfer) predicts crystallization at a range of sites (roof, walls, floor) and levels within the chamber (Irvine, 1970b).

(b) Several mechanisms of crystal-liquid segregation and crystal deposition have been proposed. All stem from the basic concept that the majority of magmas have convected throughout most of their crystallization history, and that crystals have, for the most part, accumulated from moving liquid. Wager (1963) visualized two main styles of moving liquid (slow continuous convection-driven, and faster-moving intermittent density currents), both with crystals settling under gravitation at velocities close to those predicted by Stoke's Law. Irvine (1980a) however, has suggested that crystal accumulation from both current-types, rather than being a process of gravitative settling, is more the result of flow-deposition (internal flow patterns within "flowing" crystal-liquid suspensions subject crystals to a downward-acting velocity component which carries them to the floor). Certain textural and structural features have been cited as criteria that crystals sank, floated or were transported to the floor: linear lamination, Brothers (1964); cross-bedding and slumping, Wager and Deer (1939), Thompson and Patrick (1968).

However, many features are at best ambiguous and others appear inconsistent with the "cumulus theory" (see reviews by Campbell, 1978; McBirney and Noyes, 1979). Basic inconsistencies in the concept have become increasingly evident from studies of density and viscosity relations, and rheological properties of fractionating magmas (Murase and McBirney, 1973; McBirney and Noyes, 1979; Irvine, 1980a), and this has led several workers (McBirney and Noyes, 1979; Morse, 1979) to postulate a very different process. The controlling mechanism of igneous layering (large-and small-scale) in many intrusions may not be crystal settling, but an oscillatory, diffusion-controlled process of in situ crystallization resulting from differing rates of nucleation and crystal growth (Wager, 1959; Kanaris-Sotiriou, 1974; Maaloe, 1978; Rice, 1978; McBirney and Noyes, 1979).

2.1.2 Terminology

2.1.2.1 Rock names: Despite attempts to produce a standard nomenclature for layered igneous rocks (Wager *et al.*, 1960; Wager and Brown, 1968; Jackson, 1970), no single system has been fully accepted. Terms range from descriptive (e.g. comb layering) to geographic (e.g. Willow Lake layering) to genetic (e.g. adcumulate). The terms "cumulus", "postcumulus" and "intercumulus" are firmly established in the literature. Although they presuppose some knowledge of the origin of the rocks, and their continued use has been questioned (McBirney and Noyes, 1979), the latter two terms are used in the sense defined by Jackson (1970). The term "cumulate" is used in a purely descriptive sense for igneous rocks formed by crystal accumulation; its use does not necessarily imply the gravitative settling of crystals.

There are two main approaches to naming layered igneous rocks (i) a modal classification based on all minerals, whether cumulus or postcumulus, (ii) assuming a distinction can be made between cumulus and postcumulus minerals (see section 2.1.2.2) rocks are referred to as cumulates, with the appropriate cumulus minerals as descriptors (listed in order of decreasing abundance), e.g. plag-olivine cumulate (Jackson, 1967; Wager, 1968). In this study method (ii) is adopted.

In his account of the Rhum ultrabasic sequence Harker (1908a, p. 70, 71) recognized several "major" rock types, namely "true peridotites", "anorthite-peridotites" and "allivalites". The terms "allivalite" (a plagioclase-olivine cumulate) and "harrisite" (a unique textural variety of peridotite) are retained. The term "peridotite" as applied by Harker and subsequent workers in Rhum, carries a much wider connotation than is warranted by common usage; e.g. to many petrologists the term specifically denotes a group of olivine-rich ultramafic rocks with variable amounts of Ca-poor or Ca-rich pyroxene,

and only accessory plagioclase (cf. Streckeisen, 1973). The term is applied to Rhum rocks in which the cumulus olivine to feldspar ratio is >50%. In view of the little provision for specific terminology (devoid of textural or genetic implications) of the important Fo-rich olivine-calcic plagioclase (>An_{80}) assemblages which link essentially monominerallic dunites on the one hand and anorthosites on the other, the following scheme is proposed.

(i) rocks composed of >95% cumulus plagioclase are anorthosites: plagioclase-olivine cumulates in which the two minerals are in equal proportions or plagioclase is dominant are "allivalites", i.e. plag is <95>50%.

(ii) olivine cumulates are "peridotites": those containing up to 5% postcumulus plag (±cpx) are "dunites"; variants with 5-30% and 30-50% postcumulus feldspar are "peridotites" and "feldspathic peridotites" respectively.

(iii) olivine-plagioclase cumulates are found only in the Central Series.

(iv) when clinopyroxene appears as a cumulus phase the resultant rocks are prefixed e.g. plag-cpx cumulate. This procedure eliminates difficulties arising when the rocks acquire the modal character of gabbros. Criteria for distinguishing "gabbroic" cumulates and non-layered gabbros are discussed in Chapter 3.

The above definitions are based solely on modal compositions. Textural variants are discussed in section 2.1.2.2. Clinopyroxene, although an additional subsidiary (often ubiquitous) postcumulus phase in the majority of cumulates (<5% - ca. 40 modal %), is omitted from the classification: its incorporation would unnecessarily confuse the nomenclature.

2.1.2.2 Textures: The textures of rocks from large layered basic intrusions have been described and classified by Wager *et al.* (1960). This widely accepted scheme is inherently a genetic one, based on the implicit assumption that at least some proportion of the crystals accumulated by mechanical (gravitative) segregation. The distinctions between "ortho", "meso" and "ad" cumulates are both descriptive and interpretive, and since the latter is often the primary criterion for classification, these names do not denote the geometric form of the crystals so much as the proportions of "trapped liquid". The problem of discussing the genetic aspects of igneous textures in terms of names that imply a particular mechanism of nucleation and crystal growth, is avoided by using these terms in their strict textural sense without genetic implications: viz. the textures illustrate different degrees of equilibration during crystal growth, regardless of where the crystals nucleated, and do not imply that some crystals formed elsewhere and reached their present site by sedimentation. However, any discussion of the process of nucleation and crystal growth must deal with the basic concept of cumulus theory (section 2.1.1).

Several unique textural varieties of olivine cumulate have been described from the Rhum intrusion. Peridotite (and feldspathic peridotite) containing large elongate and branching (skeletal) olivine crystals (of coarser grain-size than non-skeletal olivine in contiguous layers) are termed "harrisitic" or "harrisite" (Harker, 1908a; Donaldson, 1975a, b). Harrisites containing elongate olivine oriented perpendicular to layer boundaries are distinguished as "comb-layered harrisite" (this purely descriptive, non-genetic term (Donaldson, 1977a) is used in preference to other frequently used terms which have genetic connotations, e.g. "crescumulate" (Wadsworth, 1961) and Willow Lake layering (Taubeneck and Poldervaart, 1960)). Feldspathic peridotite

showing massive (diameter 0.15-1 m) radial growths of plagioclase poikilitically enclosing oriented subhedral olivines, is said to have a "poikilo-macrospherulitic feldspar" texture (Donaldson *et al.*, 1973). Several subvarieties of these metastable olivine and plagioclase growth patterns, used as descriptive field terms, are defined in subsequent sections.

2.1.2.3 Stratigraphic nomenclature: An informal stratigraphic nomenclature to describe the internally-conformable layers has developed from the work of Brown (1956), Wadsworth (1961) and Jackson (1970), to that adopted here: the mappable cyclic layers ("units") are based on the proportions of the cumulus phases, e.g. Units 1-15 (Brown, 1956). Groups of similar units compose a "Layered Series", e.g. the Eastern Layered Series. When a layered sequence comprises a number of non-cyclic, conformable and unconformable, macro-layers, the informal subdivision is a "Member", e.g. the Dornabac Member of the Central Series (section 2.3 this study). The complex as a whole will be referred to as the layered ultrabasic complex (L.U.C.).

2.2 Introduction to the Rhum Ultrabasic Complex

2.2.1 Previous investigation

Modern geologic mapping of the Rhum ultrabasic rocks was initiated in the mid 1950's. In the vicinity of Hallival and Askival, Brown (1956) distinguished a 470 m-thick "layered series" (hereafter named the Eastern Layered Series, E.L.S.) composed of 15 major rhythmic units (1-15) of varying thickness. Each consists of a thick olivine cumulate member (peridotite) and a thinner overlying plagioclase-olivine cumulate (allivalite). The units are locally interrupted by minor cross-cutting irregular sheets or veins of gabbroic rock (Chapter 3).

In subsequent mapping of the western portion of the complex, Wadsworth (1961) distinguished a ca. 1430 m-thick Western Layered Series (W.L.S.) consisting of four major lithological groups (unfortunately also termed "series": hereafter they will be referred to as members). These are the Harris Bay Member (ol-plag-cpx cumulates), overlain by the ol- and ol-plag cumulates of the Ard Mheall Member, which in turn are separated by a zone of "ultrabasic breccia" (defined in section 2.6.2) from the overlying Dornabac Member (plag-ol and ol-plag±cpx cumulates; and the uppermost Ruinsival Member of ol- and very minor ol-plag cumulates (lower and upper sequences are separated by a second transgressive igneous breccia). For reasons discussed later, the Lower Ruinsival and Upper Ruinsival Members are renamed the Long Loch and Ruinsival Members respectively.

Although the W.L.S. cumulates have textural and mineralogical affinities with the E.L.S., they show several significant differences.

(i) the average thickness of units is very different in each area: 350 m in W.L.S., 40 m in E.L.S.

(ii) thick plag-rich cumulates, analagous to the allivalitic portions of the 15 E.L.S. Units, are not developed in the W.L.S.

(iii) the remarkable harrisitic and comb-layered harrisitic layers, so characteristic of W.L.S. olivine cumulates (Harris Bay and Ard Mheall Members particularly) are much less pronounced in the peridotites and feldspathic peridotites of the E.L.S.

(iv) the W.L.S. contains several transgressive igneous breccia zones: the E.L.S. does not.

The different characteristics and unique distribution of these two layered series pose a number of petrogenetic problems: (a) how are the two sequences structurally related?; (b) can they be related to one common parent magma composition?; (c) what conditions of

crystallization give rise to the striking differences in their lithologies and layering style?

With regard to question (a) (and bearing in mind that the complex has been emplaced as an essentially solid mass) Wadsworth (1961) suggested that the two series were originally deposited at different levels in the original magma chamber (the W.L.S. representing a lower level than the E.L.S.), and are now exposed at the same level due to relative displacement during uplift. This movement is thought to have occurred along the Long Loch Fault. The size of the original intrusion cannot be stated with precision but it probably had much the same dimensions as the exposed portion, i.e. 7 km across.

However, preliminary mapping of the olivine cumulates immediately E and W of the fault, on eastern Ruinsival, failed to reveal any dissimilarity. Further north transgressive gabbros and igneous breccias, and later faulting have added to the complexity of the area (Wadsworth, 1961). Clearly, detailed mapping was required in this central region and on Barkeval and Trollval, to join up Brown and Wadsworth's maps (see fig 1.5 and sections 1.5.1 and 1.5.2, Chapter 1). Investigations of a more specific nature, e.g. concerning origin of the rhythmic and cryptic layering, disequilibrium textures and the igneous breccias, are summarized in Chapter 1, section 1.4.

2.2.2 Statement of the problems

The main aims of this study are (i) the development of more complex stratigraphic and petrogenetic models for the evolution of the ultrabasic intrusion (in the light of new structural, petrographical, and mineralogical data from the Barkeval-An Dornabac area), and (ii) consideration of how the Rhum magmatic system contrasts/compares with other episodes of continental magmatism that produced layered intrusions.

Since the concepts of crystal settling and cumulus processes have recently come under question (section 2.1.1) interpretation of (a) modal, textural and compositional variations (both lateral and vertical), and (b) possible origins of the rhythmic layering, must involve consideration of the wide range of possible magmatic processes which may have occurred in the chamber, viz. fractional crystallization, multiple injections of fresh magma, magma mixing, convection systems (e.g. double-diffusive), density currents, and infiltration metasomatism (section 2.1.1). These processes are interdependent, and each cannot be considered in isolation.

2.3 Summary of the Ultrabasic Sequence in the Barkeval-An Dornabac Area

(a) In the ultrabasic rocks of the Barkeval-An Dornabac area three major sequences have been distinguished on the basis of field mapping (fig 2.1). (1) the westerly continuation of the E.L.S. (units 4-12 only), (2) the northernmost extension of the Ard Mheall Member of the W.L.S.: the W.L.S. is redefined (this study) as consisting of the Harris Bay and Ard Mheall Members only, (3) a newly-defined Central Series (C.S.) composed of four members: the Outer Breccia, Dornabac, Long Loch (previously the Lower Ruinsival Series), and Ruinsival (previously the Upper Ruinsival Series) Members. All four were previously regarded as part of the W.L.S. (Wadsworth, 1961).

(b) The Central Series (not referred to as "Layered" since the majority of Members lack well-developed fine-or medium-scale rhythmic layering) is transgressive to the E.L.S. and W.L.S. At its northern and southern extremities, it extends across the Inner Ring Fault (fig 2.1) and must therefore have formed after emplacement of the E.L.S. and W.L.S.. The C.S. is believed to have crystallized in situ.

(c) The structural relationship of the E.L.S. and W.L.S. is unresolved.

(d) The striking differences between the three series are inferred to reflect a fundamental difference in the way they accumulated and solidified.

(e) The significant role played by the Long Loch Fault in the development of the C.S., and the relevance of this Series to the evolution of the complex could not be appreciated by early workers because of the lack of detailed mapping of the central region. It is clear that the new stratigraphic, petrologic and structural data require a more complicated interpretation (see relevant sections).

2.4 The Eastern Layered Series (Barkeval): Field Relations

2.4.1 General introduction

The E.L.S. outcrops for a further 2.5 km westwards from the Hallival-Askival "type-area", before being truncated by the C.S. Exposures in the Barkeval area have been mapped in detail (see Map 1, in the pocket at the back of Vol. 2). No other layered ultrabasic rocks are found overlying it, although several minor peridotitic intrusions (Chapter 5) and one Member of the C.S. (section 2.6) do cut the Series. The E.L.S. remains structurally intact, although minor offsets appear to have occurred along a set of nearly-vertical NW-SE, N-S and NE-SW trending faults perpendicular to the strike of the igneous layering. Within the study area the most extensive exposures of the E.L.S. are on the N and W slopes of Barkeval: abundant faulting is responsible for the observed discontinuities along strike, while much detail is obscured by moraine.

Three stratigraphic traverses (fig 2.2 and Map 2) show an estimated maximum thickness of ca. 290 m: only Units 4-12 are exposed.

Units were mapped using the easily-traceable allivalite scarps, and individual units distinguished on the basis of persistent marker horizons, and lithological and structural differences. Lateral changes in strike and dip of layering, and in unit thicknesses are minimal. This section deals with vertical and lateral variations, including field characteristics, internal structures, style and types of igneous layering. Stratigraphic changes in mineralogy and textures are discussed in section 2.7

2.4.2 Unit 4

Unit 4, the lowest on Barkeval, is exposed only in the extreme NW corner of the E.L.S. (300 m south of Priomh Lochs). Two thin plag-ol cumulate horizons (8 m apart and with a maximum strike-length of 300 m) mark the top of the unit. The lower of these allivalite horizons (4 m thick) shows a complex modal variation in proportions of cumulus olivine and feldspar: a 75 cm basal sequence of finely-alternating olivine and plag-ol layers passes sharply upward into 60 cm of fine-grained peridotite. This in turn is overlain by 1 m of plag-ol cumulates (highly-slumped in the upper 50 cm), 60 cm of disturbed olivine cumulate, and finally 2 m of layered allivalite (10 cm layers defined by olivine concentrations). The upper contact is exposed and no chromitite layer is developed.

The more uniform upper allivalite horizon (4 m) consists of non-graded 10-20 cm layers defined by varying olivine:plagioclase (cumulus) ratios. Upper and lower contacts are not exposed.

2.4.3 Unit 5

This unit has a maximum thickness of 37 m (fig 2.2). The thick sequence of olivine cumulates shows well-developed fine-scale layering (10-30 cm scale) defined by sudden changes in olivine grain-size (e.g. 0.5→2 mm), individual layers being non-graded. Some show planar

lamination (parallelism of elongate olivines), whilst others are rich (>3%) in disseminated spinel. Poikilitic clinopyroxene (1-4 cm diameter) is widespread, giving rise to characteristic pitted (carious) surfaces.

The thin allivalite maintains a constant thickness along strike (3 m) and upper and lower contacts are sharp (undulatory and flat respectively). The cumulate is isomodal, lacks fine-scale layering, and contains a distinctive marker horizon in the form of a 0.1-0.5 m thick coarse-grained peridotite with sharp irregular contacts (the immediate underlying 50 cm of allivalite are disturbed, fig 2.3). A thin impersistent 1-3 mm spinel cumulate (chromitite) marks the top contact of the unit.

2.4.4 Unit 6

The peridotite is similar to that in Unit 5 and maintains a constant thickness along strike (20 m). In contrast the overlying allivalite thins markedly from at least 4 m in Coire Dubh (the base is unexposed), and shows a number of distinctive features. (a) large (up to 4 mm) anhedral cumulus olivines are partly poikilitic towards plagioclase laths, and impart a very coarse-grained appearance to these rocks: the feldspars show a pronounced planar lamination; (b) thin single or multiple peridotite layers, characteristic of Units 4 and 5 are absent: the allivalite is, in lower levels at least, isomodal; (c) much of the uppermost "rubbly-weathering" 1.5 m is rich in large (2 cm) green poikilitic clinopyroxenes, whose modal proportions are highly-variable (av. 5-30%) over a few cm and define a wispy, streaky "layering". This pyroxene-rich variant disappears westwards; in the extreme west only the laminated isomodal allivalite is present; (d) 3 x 12 cm wedges and lenses of anorthosite are present in the topmost 0.5 m.

As in Unit 5 the base of the allivalite is very irregular (wavelength 0.2-0.5 m), and a thin (2-3 mm) chromitite is intermittently developed at the upper contact.

2.4.5 Unit 7

This unit, the most clearly defined along the foot of the northern slopes of Barkeval (fig 2.4), has a well-defined leucocratic section (maximum 5 m thick). As with the majority of plag-ol cumulates, the base is sharp and highly sinuous, and the rock is isomodal (without fine-scale cumulus layering). The distinguishing features are:

(a) the disturbed nature of the lamination, and the presence of several 10 x 30 cm blocks of similar, but undisturbed, material and numerous smaller (2 x 5 cm) ellipsoidal inclusions of anorthosite. The degree of disturbance increases westwards to that shown in fig 2.5.

(b) an uppermost 20 cm consisting of well-laminated, undisturbed, more feldspathic material with single or multiple spinel seams: the exact nature of this complex zone varies along strike (fig 2.6). The lower chromitite (<1 mm thick) marks the base of the anorthositic layer, and is not always present. The middle seam (also intermittently present) is only two or three crystals thick, undulatory, and may or may not be overlain by a thin (7 mm) fine-grained olivine cumulate. The well-developed upper seam is the most persistent and varies from 2-15 mm thick. When of uniform thickness it is characteristically "dimpled" (wavelength 1-8 cm, amplitude 1-4 cm); in other cases the base can be very irregular and the top planar.

In the more disturbed western exposures the lower and middle seams are absent. The distribution and conditions of formation of the spinel layers are discussed in section 2.7.4.5.

In the underlying olivine cumulates layering is, as in other units,

defined by grain-size variations. However, layers average >1 m, can be size-graded (fining upwards) and show transitional contacts marked by 2-4 cm of laminated elongate olivines. In addition, the coarser-grained cumulates tend to be dominant: those with olivines >4 mm (and a tendency for elongation) are termed "protoharrisite" (an incipient form of harrisite).

2.4.6 Unit 8

The Unit 8 allivalite forms the most prominent scarp in Coire Dubh (fig 2.4). The lower half shows well-developed lamination and fine-scale layering. A 3-4 mm spinel-olivine cumulate marks the base. The upper 6-8 m are characterized by a marked increase in the amount of clinopyroxene, and a pseudo-coarse-grained appearance (cf. features (a)-(d) in Unit 6). Cyclic layering (0.5-1 m scale) involves olivine-rich layers (with a high % of poikilitic pyroxene, whose diameter increases upwards) sharply overlain by finely-layered cpx-plag cumulates. Contacts between types are sharp and irregular. Contacts with overlying Unit 9 rocks are not exposed. In the thinner western exposures (maximum 5 m) the pyroxene-rich facies is not developed and the upper contact is sharp and planar (fig 2.7).

The Unit 8 olivine cumulates show little variation from the sequences described from previous units.

2.4.7 Unit 9

The lowermost plag-ol cumulates have affinities with those of Unit 7, and the lower contact is transitional: over a 2 m zone several minor (5-10 cm) cumulus plagioclase-rich layers alternate with peridotite (protoharrisite and "normal"). Cumulus clinopyroxene makes a sudden appearance in the upper 2 or 3 m (cf. Unit 8), and cpx-plag cumulates (10-25 cm) alternate with plag-ol cumulates. No chromitite is developed at the upper contact. The underlying olivine cumulates are finely-layered in the style of Units 4 and 5.

2.4.8 Units 10-12

The leucocratic cumulates of these three units are each characterized by a complex combination and intercalation of peridotite and allivalite, and transitional contacts. The most continuously exposed sections are (a) on northern Barkeval, in a faulted cliff face 350 m SSE of Triangular Loch (G Ref 378975), and (b) G Ref 374976, on Barkeval's western face.

A very distinctive "clotted" texture in the basal rocks of Unit 10 allivalite heralds the incoming of cumulus plagioclase (fig 2.8). The 20 m-thick allivalite (fig 2.9) consists of alternating zones of slumped (disturbed) and undisturbed laminated plag-ol cumulates (each 3-4 m) plus an occasional 10 cm peridotite layer, and grades by an increase in the peridotite/allivalite ratio into Unit 11 (chromitite is absent). As in Units 7 and 9, anorthosite lenses are commonly associated with disturbance. In eastern exposures only (Coire Dubh), the allivalite shows affinities with Units 8 and 9, namely the presence of large anhedral olivines, and sudden incoming of abundant clinopyroxene (both cumulus and intercumulus).

The Unit 11 plag-ol cumulates (12 m) lack slumped horizons, but contain more numerous multiple concentrations of cumulus olivine, averaging 5-10 cm each and concentrated into at least five specific levels. One peridotite layer, 1 m from the top, has a distinctive undulatory upper contact (fig 2.10a). The fact that the laminar fabric in the leucocratic cumulate continues undisturbed through the projections (fig 2.10b) is suggestive of a "replacement" origin (similar to the large dunite pipes replacing harzburgite in the Duke Island, Stillwater+Bushveld intrusions ; Hess, 1960; Cameron and Desborough, 1964; Irvine, 1980)). The Unit 11-Unit 12 contact is marked by a relatively undisturbed chromitite (3 mm-1 cm thick) overlying a 10 cm anorthosite layer.

Unit 12 allivalite is instantly recognizable by the development, within its 15 m section, of five evenly-spaced 0.5 m-thick peridotites (each with a flat base and undulatory top). The intervening plag-ol cumulates are undisturbed, free of inclusions, and weakly-laminated and -layered.

2.5 The Western Layered Series : Field Relations

Only the uppermost (Ard Mheall) member of the W.L.S. (as defined in section 2.3) is present in the study area. Exposures extend northwards from Loch an Dornabac (the northern limit of Wadsworth's (1961) map) for another 0.5 km until, in the N and E, the Ard Mheall sequence is truncated by the C.S. The rocks are predominantly olivine cumulates, with minor ol-plag cumulates (spinel is a ubiquitous minor cumulus phase). Regular rhythmic layering, well-developed throughout, dips generally E or SE, and increases from the more usual 15-20° to ca. 30-40° (and as much as 60°) towards the contact with the C.S. Three styles of layering are present:

(a) in the lower half of the exposed succession thick 1-2 m layers of harrisitic cumulates (with flat basal, and irregular upper surfaces) alternate with thinner, less abundant, non-harrisitic rocks.

(b) in the upper half of the sequence, harrisite is a less abundant variant (<25% of the total sequence). Here the fine-scale layering (on scale of ca. 10 cms) involves variation in olivine grain-size (and hence % of intercumulus silicates) such that fine-grained dunite alternates with coarser-grained peridotite or feldspathic peridotite. Thin continuous, closely-spaced, chromitite layers (2-5 mm) are numerous.

(c) cumulus plagioclase appears in the topmost exposures (ca. 6 m): on a scale of 2-5 cm, olivine cumulate layers alternate with anorthosite. Both olivine and plagioclase show a planar lamination.

Layers are neither grain-size nor modally graded (section 2.7).

2.6 The Central Series : Field Relations

2.6.1 Introduction

The Central Series takes the form of a large (8 km N-S) dyke-like mass, ranging in width from 0.5 km in the north (Minishal) to 2 km at the south coast (fig 2.1), and with a maximum exposed thickness of ca. 900 m. In fig 2.1 the northern half of the C.S. has been mapped in detail (this study, Map 1): data for the area south of Loch an Dornabac is taken from Wadsworth (1961). Contacts with the E.L.S. and W.L.S. are of two main types; discordant intrusive (with or without the development of an igneous breccia) and faulted. The four constituent elements of the Central Series are, in order of formation, the Outer Breccia, Dornabac, Long Loch and Ruinsival Members.

2.6.2 The Outer Breccia Member

2.6.2.1 Description: Of the five northerly-striking discordant igneous breccia bodies (each consisting of blocks demonstrably derived from the layered series, enclosed in a feldspathic peridotite matrix texturally similar to the layered series rocks; and with a ratio of blocks:matrix >3:1) in central and SW Rhum (Wadsworth, 1961; Donaldson, 1975b), the Outer Breccia is the largest. Its westernmost exposures are equivalent to, and continuous with, the Lag Sleitir breccia (Wadsworth, 1961). Exposure is moderate (west of the Long Loch Fault) to excellent (east of the fault). Contacts with the E.L.S. and W.L.S. rocks are exposed over 80% of their strike-length, vertical, relatively sharp (being transitional over 1-2 m), and defined by the first appearance of intense veining by feldspathic peridotite (vein: host rock >1:2). Country rocks have neither slumped nor mixed with the matrix, suggesting they deformed brittly during breccia formation.

Lack of matrix chills and thermal metamorphic effects suggest the layered series were still hot.

The breccia is remarkably heterogeneous with respect to lithology, textures, dimensions and "density" of the blocks, and the texture of the matrix. Blocks range from fine-grained dunite (fig 2.11a) to coarser-grained normal and harrisitic peridotite (fig 2.11b) and rare allivalite; in size from <10 cm to >2m (fig 2.12); in shape from rounded (fig 2.11a) to angular (fig 2.12); and may be layered (fig 2.11b) or unlayered (fig 2.11a). Layered blocks show a random orientation. Nearest the outer contacts blocks tend to be smaller, more rounded, more closely packed, and locally derived (the dominant block-type is directly related to the adjacent layered series rock type: viz. abundant allivalitic blocks are found immediately adjacent to in situ E.L.S. allivalites).

The peridotitic matrix is equally heterogeneous, varying (within a few cms) in olivine grain-size (<1-3 mm), percentage of intercumulus feldspar (10-50%) and surface texture (uniform - fig 2.11a - to poikilo-macrospherulitic - fig 2.13). The block: matrix ratio randomly varies from <0.1- ca. 0.8 (av. 0.5-0.7). Both blocks and matrix are often cross-cut by gabbroic veins (0.4-1 cm wide) of plag-cpx-ol material (?filter-pressed).

SE of Loch an Dornabac, and 200 m E of the Long Loch Fault, the upper 10 m of breccia contain several prominent harrisitic layers, 0.5-3.5 m thick, dipping <18°, with well-defined sharp boundaries.

2.6.2.2 Discussion: Two contrasting origins have been postulated for the Rhum igneous breccias. Wadsworth (1961, 1973) regarded them as fault scarp scree-deposits, formed during differential movements on the reservoir floor (blocks = brecciated consolidated cumulates, matrix = contemporaneous infilling of interstices by unconsolidated

primary cumulates). Donaldson (1975b) favoured an intrusive origin, with the breccias representing either (a) filled conduits through which magma (carrying a considerable proportion of suspended olivine crystals) was supplied to the main chamber, or (b) rapidly-mobilized "pods" of magma (being pockets of "uncrystallized" liquid, trapped within the cumulate pile). Field evidence presented above (and in sections 2.6.4 and 2.6.5) favours an intrusive origin. The various stages of brecciation envisaged are illustrated in fig 2.14.

2.6.3 The Dornabac Member

2.6.3.1 Description: Rocks of the markedly feldspathic Dornabac Member (dominantly gabbroic cumulates, i.e. plag, ol, cpx) overlie the ultramafic breccia. The contact is sharp, defined by the first appearance of abundant cumulus feldspar (± clinopyroxene) and the development of fine-scale rhythmic layering, and is exposed for ca. 50% of its strike-length. The most important aspect of its distribution is that the Dornabac Member also occurs to the east of the Long Loch Fault (Map 1 and fig 2.1). West of the fault the outcrop covers 2.5 km from An Dornabac ridge to the Stable Flats area; only 700 m of strike-length are exposed intermittently to the east.

The Dornabac Member reaches its maximum stratigraphic thickness (ca. 150 m) to the east of Loch an Dornabac, and is characterized by alternating layers of ol-plag-cpx, cpx-plag-ol and plag-ol cumulate which dip regularly eastwards at 30-40° (fig 2.15) and are highly disturbed (with evidence of slumping), irregular and impersistent. The top 50 m (missing in the An Dornabac ridge due to a transgressive gabbro intrusion) are exposed only on a small ridge NE of the loch. The sequence consists of 25 m of highly-feldspathic ol- and minor plag-ol cumulates which are heterogeneous with respect to relative proportions of cumulus olivine and feldspar, olivine grain-size, and percentage of

intercumulus feldspar and pyroxene (fig 2.16). Small-scale (10 cm) spherulitic intercumulus feldspar growths are common ("coralline" or "bird-track" texture), and the rocks often show evidence of high-temperature shearing (planes marked by concentrations of feldspar - see fig 2.16).

The upper 25 m (a series of small gently-dipping escarpments) display a highly-distinctive poikilo-macrospherulitic texture (defined in section 2.1.2.2). Feldspar "rays" can (a) radiate from a sharp or diffuse nucleus, and show open or closed branching (fig 2.17a), (b) show partial radiate structure (fig 2.17b): two bundles of oppositely directed rays define a "bow-tie" type (Lofgren, 1974), (c) be small-scale (<10 cm), irregular and lack extensive branching: this incipient or "juvenile" type is termed "coralline" or "bird-track" (this study). All morphological types are intergradational and randomly distributed.

Intercumulus layering (defined by the relative abundance of intercumulus feldspar and pyroxene) is discontinuous (maximum strike-length 10 m), but conformable with the main Dornabac cumulus layering (av. 35°). Diffuse patches and veins of anorthositic gabbro (similar to those described by Donaldson, 1975b), are present.

The Dornabac Member thins rapidly northwards (only 10 m are present in the Stable Flats, pyroxene-rich cumulates only) and eastwards (35 m found east of the Long Loch Fault, and the poikilo-macrospherulitic facies is absent). Lithologies, layering-style, and textures in the eastern exposures, are identical to those in the type-area, but dips are considerably steeper (50-60° due west). Many exposures show high-temperature drag- faulting (fig 2.18).

2.6.3.2 Discussion: Both field relationships and petrography suggest that the Dornabac Member accumulated soon after the formation of the

Outer Breccia, during a continuing period of tectonic disturbance (hence widespread disruption, slumping and drag-faulting of semi-consolidated layers). The steep dips (>40°) do not represent original angles of deposition, and are regarded as being the result of tilting (?due to differential subsidence). The distinctive topmost 50 m sequence (and its restricted distribution) may represent emplacement of a separate magma portion (see section 2.7.3.9).

2.6.4 The Long Loch Member

Previously defined as the Lower Ruinsival Series (Wadsworth, 1961) this sequence has been renamed because (a) its geographical distribution extends far beyond the Ruinsival area, and is more closely linked to the Long Loch Fault, and (b) several significant field characteristics (in the An Dornabac-Barkeval area at least) distinguish it from the Upper Ruinsival Series.

The Long Loch Member conformably overlies the Dornabac Member, overlaps southwards onto the Outer Breccia and W.L.S. (fig 2.1), and extends for 8 km over the entire length of the complex. The maximum stratigraphic thickness (460 m) is in the Glen Harris area: approximately 200 m are exposed in the study area. Like the Dornabac Member, this sequence is also found east of the Long Loch Fault.

In the type-sequence (SE of Loch an Dornabac) the basal contact is sharp and marked by a sudden return to massive olivine-rich (dunitic) cumulates. The first few tens of metres consist of fine-grained dunite. Olivine grain-size, morphological and modal variations are negligible, and sporadically-developed incipient igneous layering (dipping 15-20° towards the Long Loch Fault) is principally defined by (a) multiple disseminated-spinel concentrations (i.e. spinel-ol cumulates) in the form of impersistent layers (ranging 2 mm-25 cm thick), and (b) orientation of wedge and clot-like concentrations of intercumulus feldspar (fig 2.19).

These rocks are overlain by approximately 5 m of disrupted and fragmented layers (5-10 cm) of coarse-grained plag-ol cumulate (fig 2.19), followed by ca. 10 m of massive dunite containing horizons rich in large angular allivalitic "wedges".

The remainder of the sequence (towards the Long Loch Fault) comprises heterogeneous feldspathic olivine cumulates showing (1) randomly-mixed olivine grain-sizes (<1 mm to >1 cm), (2) short discontinuous chromitite seams, (3) streaky intercumulus layering; the increase in dip from 25° to nearly vertical (adjacent to the fault) defines an overall "wedge-shaped" geometry for the Member. In addition, several breccia zones are developed, increasing in prominence towards the fault. A similar, but condensed, sequence is found east of the fault.

The salient features of the Long Loch Member are therefore (a) predominance of olivine-rich cumulates: plagioclase is rarely a cumulus phase, (b) lack of fine-scale rhythmic layering (found in the E.L.S. and W.L.S. peridotites), (c) absence of skeletal olivine morphologies and "harrisitic" texture.

A distinctive layered sequence found in the Stable Flats - Minishal area (the northern "prong" of the complex) is regarded as a lateral variation of the Long Loch Member (fig 2.1 and Map 1). The rocks outcrop in a basin to the west of Long Loch, and layering defines a shallow asymmetrical synform with eastern and western limbs dipping at 20° and 30° respectively. Three conformable units are present, best exposed on the ridge along the west shore of the loch. The lowest, unit A, consists (south of the road, G Ref 363992) of twelve layers of careous-weathering olivine cumulates; the lower ten (each ca. 6 m thick) show a fining upwards of olivine grain-size (4 mm → ½ mm). Layer 11, a 2 m harrisite, is overlain by a 2 m sequence of fine-scale rhythmically-layered fine-grained peridotite (Layer 12).

Unit B (6.5 m thick) comprises ten layers of ol,plag cumulate (fig 2.20) ranging in thickness from 0.2-1 m, and each graded with respect to the amount of intercumulus clinopyroxene (decreases upwards). Some layers have irregular bases and cut down into underlying layers; others (especially near the base of the unit) are rich in 10-20 cm subrounded allivalite fragments. The base of Unit B is poorly exposed, but sharp and highly irregular (wavelength 30 cm, amplitude 4-8 cm). Several olivine-rich pipe-like projections, found at the base of some layers, appear to cut the igneous lamination and are regarded as "replacement" structures (cf. fig 2.10b).

Unit C consists of one continuously-graded olivine cumulate layer (ca. 75 m). The base is sharp and defined by numerous small subrounded to subangular olivine clusters (non-touching) set in a feldspar-pyroxene "matrix" (fig 2.21); occasional peridotite and allivalite xenoliths are present. Within 2 m of the base, the clusters fine upwards into single crystals (av. 2 mm) in a feldspathic peridotite. Approximately 50 m from the base subsidiary cumulus plagioclase appears. The size of olivine clusters, together with the size and abundance of rock fragments, decrease northwards, and in several places Unit C peridotite has wholly or partially "peeled off" top layers of the Unit B allivalite. These features suggest magma (rich in suspended olivine aggregates) moved northwards along the contact, possibly in the form of a suspension-type density or turbidity current.

Units A-C are regarded as having formed from pulses of "Long Loch Member" magma which spread laterally over a narrow shelf-area (the units thereby forming a tabular body). Unit C grades southwards into the Long Loch Member proper. Unit B (eastern limb only) can be traced southwards for 1 km, to the east of the Long Loch Fault, but closes northwards, approximately 100 m north of the road (Map 1). Unit A

cannot be traced south of Long Loch, but appears to be continuous to the north, with the heterogeneous non-layered peridotite of the main Minishal mass.

2.6.5 The Ruinsival Member

The Ruinsival Member (formerly the Upper Ruinsival Series; Wadsworth, 1961) extends from Ruinsival (the type-area) across Glen Harris to the northern slopes of Barkeval (Map 1). Exposure is excellent and the maximum stratigraphic thickness, in the study area, is ca. 200 m (310 m on Ruinsival). In contrast to the Dornabac and Long Loch Members, these olivine cumulates form a large, ?single, intrusive body, with steep transgressive contacts against the E.L.S. and the Outer Breccia, Dornabac and Long Loch Members. The rocks are not unlike the dominant lithology in the Long Loch Member; viz. a thick sequence of unlayered peridotite, heterogeneous (over an area of a few cm or tens of metres) with respect to (i) texture (smooth, fig 2.22, to finely-pitted, fig 2.23, to poikilo-macrospherulitic and coralline, fig 2.24: harrisitic type is absent) and (ii) percentage of intercumulus silicates (compare figs 2.22, 2.23 and 2.25). A few minor patches (4 m^2) contain subrounded dunite blocks (maximum 0.5 diameter).

The peridotite mass shows a variety of contact relationships with the host E.L.S. and C.S. rocks; vertical/moderately-dipping, faulted or intrusive (with or without the development of igneous breccia). The NW contact (377975) is approximately vertical and marked in part by an intrusive gabbro. Southeastern contacts (378972 and 381969) are faulted; the former is marked by a distinctive near-vertical gully which bisects the southern face of Barkeval. The western contact dips ca. 45°E and over at least 50% of its length is marked by an igneous breccia (best developed against the E.L.S., fig 2.26). Against the C.S. in Glen Harris, the breccia is less spectacular.

2.7 Petrography

Lithologies are highly-variable: a general idea of the abundance, variety and distribution of the rocks may be seen in the sections given in Tables C.1 + C.2 They summarize, in table form, the petrographic and mineralogic variety observed in the layered sequence. During study of the E.L.S. and C.S., specific features were selected in anticipation that they would offer means of stratigraphic control. Selection was based on the assumption that the feature was controlled by a process which operated over the whole magma chamber at, or nearly at, the same time. Size variation of cumulus phases and modal variations (plus compositional variations, sections 2.9 and 2.10) were examined.

2.7.1 Mineral proportions

Without details of the stratigraphic sequence and modal abundances within the lithological units making up the E.L.S. and C.S., attempts to model the petrogenesis of the complex are meaningless; even on the relatively small scale chosen to represent lithological variations (samples every 3 m where possible), many finer-scale and possibly significant details cannot be shown. An adequate explanation of the modal data will require (i) a comprehensive analysis of layering features, (ii) extensive data on mineral compositions, and (iii) knowledge of the dynamics of flow of fluids in systems of this type (Irvine's 1980a paper represents the first comprehensive attempt at understanding these processes).

In measurement of modes it was found impractical to determine the amount of postcumulus overgrowth on cumulus crystals. Such material is counted as part of the cumulus mineral (this undoubtedly gives rise to an overestimation of the volume of cumulus minerals, but probably does not have an appreciable effect on their relative

proportions). The standard technique of modal analysis by point-counting thin sections is useful only for those samples that are representative of relatively thick, uniform (isomodal) layers. In cases where samples have large poikilitic grains or low concentrations of a particular cumulus mineral, modal proportions have been estimated in the field.

The northern and western logged sections on Barkeval (located on Map 2) were chosen where the sequence is best exposed and the location of, and displacement on, faults is well known. The Central Series has been preliminarily sampled in a W-E traverse, immediately south of Long Loch (Map 2).

Over 130 samples were collected, sectioned and examined petrographically. Modal data are presented in Appendix C, and illustrated in figs 2.27 and 2.28.

In fig 2.27 systematic variations are immediately obvious. Olivine (a cumulus phase throughout the series) contents (60-70%) at the base of each peridotitic sequence are similar for successive units, decreasing slightly to ca. 50% immediately below the allivalitic horizons. The changeover from peridotite to allivalite is abrupt and marked by a sudden decrease in olivine-content. Olivine decreases throughout the plagioclase cumulates to less than 20% at the tops of the cyclic units. Units 11 and 12 allivalite are atypical in that the former shows an initial marked increase in olivine-content at the base (30-65%), and the latter a steady upward increase in olivine-content throughout (20-40%).

Cumulus plagioclase makes a sudden appearance at the base of each allivalite (45-55% modal range), and increases to a maximum of 72% in the olivine-bearing layers and 99% in the anorthosites. Contents of intercumulus feldspar show little variation throughout the olivine cumulates (generally 20-40%).

Intercumulus clinopyroxene contents are generally constant throughout the units and, if anything, slightly lower in the allivalites. Cumulus grains make a sudden appearance in the upper third of Unit 8 and 10 allivalites, and average 45%.

The modes of individual samples are plotted in terms of ol-plag-cpx in fig 2.28. The E.L.S. data show a distinct bimodality, with allivalites and peridotites forming separate non-overlapping data clusters, and the former extending towards the clinopyroxene apex with the incoming of cumulus pyroxene.

Central Series cumulates show a greater range of olivine-contents, by overlapping considerably with the E.L.S. data and extending towards the olivine apex.

2.7.2 Size variation

Visual estimates of grain-size during exposure logging were classified as fine-grained (<1 mm), medium-grained (1-3 mm) and coarse-grained (>3 mm). No systematic grain-size variations were found within cyclic units and layers of the E.L.S. or C.S., because the sampling interval (3 m) is greater than the average scale of grain-size variations (often <0.3 m). Grain-size grading of layers is largely absent (section 2.4). Attempts to correlate detailed grain-size sequences laterally over distances greater than 10 m failed, and had only limited success over shorter distances.

A second method of grain-size examination was more qualitative and allowed a check on semi-quantitative data. Length and width measurements of cumulus olivine and plagioclase were made on 500 grains per thin-section (technique and limitations of this method are discussed by Jackson, 1961, p. 21). Data are summarized in Appendix C, and there are no significant lateral variations in grain-size of either phase at any given horizon.

2.7.3 Textures

2.7.3.1 Introduction: The bewildering array of textures in the Rhum ultrabasic rocks (e.g. careous, harrisitic, coralline, poikilo-macrospherulitic, mottled) is the result of combinations of the following parameters:

(i) the variety of both cumulus and intercumulus phases

(ii) their modal proportions (see section 2.7.1)

(iii)their crystal morphologies and grain-sizes (see section 2.7.2)

(iv) presence or absence of a preferred orientation of minerals.

Textures of the Rhum cumulates have been extensively described and illustrated by Brown (1956), Wadsworth (1961) and Donaldson (1975a, b, 1977a, b), therefore only a summary of olivine, plagioclase and pyroxene habits is given here. New data are presented for the Cr-spinels, ilmenite and sulphides. Figs 2.29 to 2.30 show a representative range of textures.

2.7.3.2 Olivine: Grains are rarely euhedral, being modified by varying degrees of postcumulus overgrowth to subhedral (in the peridotites, fig 2.29) and anhedral in the orthocumulate plag-ol cumulates (margins subpoikilitic towards plagioclase - fig 2.30). In non-harrisitic olivine cumulate layers the majority of grains are equant and therefore a preferred orientation is rare. The typical fabric is a partially "grain-supported" one (fig 2.29). When grains interfere, they do so along planar or slightly curved boundaries and with grain boundary angles close to 120°. Olivines enclosed in poikilitic pyroxenes are commonly smaller and more rounded than those outside. In many samples the larger grains (>1 mm) often contain oriented platelets consisting of intergrowths of magnetite and pyroxene (products of high-temperature oxidation of olivine, Putnis, 1979) - see fig 2.31. The degree of alteration is variable - generally weak in the E.L.S., moderate in the C.S. - and products include yellow and green serpentine, chlorite, magnetite and carbonate.

2.7.3.3 Plagioclase: Feldspar is entirely intercumulus in all E.L.S. and the majority of C.S. peridotites. Grains are poikilitic towards olivine, non-clouded and twinned predominantly on the pericline law. They rarely show evidence of strain. In the plag-ol cumulates the euhedral, lath-shaped prisms commonly show a marked tendency towards planar lamination (fig 2.30). Inter-grain boundaries, as for olivine, are straight or slightly curved and occasionally weakly interpenetrant.

2.7.3.4 Clinopyroxene: Intercumulus poikilitic pyroxene is similar in both E.L.S. and C.S. - fresh, unschillerized, rarely twinned,and lacking exsolution lamellae. Cumulus pyroxene is markedly different in the two series - compare figs 2.32 and 2.33.

2.7.3.5 Cr-spinel: Cr-spinel is disseminated throughout the L.U.C. in amounts ranging from 2-5%. At numerous horizons it is concentrated (>10 modal %) into layers:(a) ol-spinel cumulates, (b) spinel-ol cumulates, (c) spinel cumulates (chromitites).

Type (a) and (b) layers range from <1 cm to >30 cm in thickness and are found, often alternating, in peridotites only. Individual layers cannot be traced for more than about 10 m in the E.L.S. and 2 m in the C.S.

Chromitite layers are most persistently and abundantly developed in the E.L.S., where they vary in thickness from 0.2 mm to 1.5 cm and can be traced for at least 2.5 km along strike. Their distribution within the E.L.S. is more complex than has been described by previous workers. They are found (i) at the base of cyclic units, between allivalite and the overlying peridotite (Brown, 1956; Henderson and Suddaby, 1971): in the Barkeval sequence spinel layers have been identified at the base of Units 6, 7, 8, 12; (ii) at the contact between olivine and plag-ol cumulates within Units 8, 11; and

(iii) within olivine cumulates, e.g. overlying harrisite layers, or more often, alternating with olivine-rich and spinel-rich layers (type (a) and (b) above) which collectively constitute a chromitite zone (Jackson, 1963). Numerous such zones ranging in thickness from 0.1-1 m have been recognized in the Barkeval sequence, and the number of chromitites in a single zone varies from one to more than 10, but is generally two to five. Persistent chromitites have yet to be found within the allivalites.

Although the spinel cumulates are persistent over large distances their detailed structures are highly variable. Fig 2.34 illustrates the variations both between and within individual layers from Units 8-12. The sharp upper and lower contacts vary from planar (M46, B78, KRG, B54) to slightly-irregular (J24) to highly-undulatory (M68/A, M167) and consequently the thicknesses vary from uniform (e.g. J24, M46, M102) to highly-variable (M68/A, M167). Although the majority of spinel seams occur at the allivalite-peridotite interfaces, in some instances they are to be found a short distance above the lithological contact (e.g. E19, E30).

Several of the spinel cumulates show signs of syn- or post-deposition deformation. During the former, the layer either remained coherent and simply "buckled" (e.g. M68/A), or lost coherency and pulled apart (e.g. E1); during post-depositional disruption the spinel layers behaved rigidly (e.g. B78/1, M46).

The chromitites consist, on average, of 50 modal % spinel in poikilitic plagioclase (and rare 1 mm poikilitic olivine grains), and show only a poor development of synneusis structure. Crystals are highly variable in both shape and size, ranging from euhedral to rounded subhedral to "hopper" type (analogous to hopper olivine morphology, Donaldson, 1976), and from 1 mm (euhedral grains) to <0.1 mm (the

rounded variety). The relative abundance of the three morphologies varies from seam to seam, but in general the euhedral and subhedral grains are in roughly equal abundance and the hopper-type subordinate. Only in one of the sampled layers was the hopper-type dominant (M167). There is little or no grain-size sorting in the majority of seams, the exceptions being J8 (fig 2.34) which consists of two upward-fining layers (a planar contact separates the two), and B78/2 which only apparently coarsens upwards, due to mutual intergrowth of crystals.

The spinels are translucent in varying shades of brown, although in sample J8 they are a distinctive olive-green. In each seam, 1-4% of crystals contain roughly spherical silicate- and/or sulphide-rich inclusions (this figure rises to ca. 30% in M167). The inclusions measure 10-30 μm in diameter and contain predominantly orthopyroxene (± biotite, amphibole and ?feldspar) in samples M167, J8, M102 and Cu-Fe sulphides in E1, B78/1/2, KRG. Similar silicate inclusions have been described from Muskox (Irvine, 1974a), Bushveld (McDonald, 1965) and Stillwater (Jackson, 1961).

At high magnifications (x 160) oil immersion reflected light microscopy revealed that the majority of Cr-spinel grains in the seams contain a high concentration of very fine oriented exsolved platelets which are just within resolution of the objective. Putnis and Price (1979) have identified two exsolved phases - a magnesian ilmenite and a metastable spinel phase - and concluded they are a result of oxidation of the Cr-spinel at moderate (>600^{o}) temperatures.

From fig 2.34 it is noticeable that all spinel layers at peridotite-over-allivalite contacts are underlain by a thin anorthosite (0.5 - 2 cm).

Disseminated spinels are found, individually or in small clusters, in five different local environments, (a) totally enclosed by olivine

in the peridotites, (b) within embayments in olivine (and sitting in plagioclase), (c) within poikilitic plagioclase, not adjacent to olivine, (d) totally enclosed by cumulus plagioclase in the allivalites, and (e) between cumulus plagioclase laths. A further description of these environments in terms of the petrography of the rocks is given in section 2.9.4.

The majority of grains contain oxidation-exsolved ilmenite (only those totally enclosed by cumulus olivine and plagioclase are consistently homogeneous), and the range of Cr spinel-ilmenite textural associations found is summarized in fig 2.35. All intergrowth types can be found in any one of environments (b) to (e).

Origin of the chromitites: The presence of small clusters of Cr-spinel between cumulus olivines, the entrapment of occasional crystals in olivine as early-formed inclusions, and the observation that the average modal ratio of Cr-spinel to olivine (1:24) is that in which picrochromite and forsterite co-precipitate (initially, since the ratio decreases with fractionation as a result of the curvature of the cotectic boundary) in the system $MgO-Cr_2O_3-SiO_2$ (Keith, 1954), suggest that the two minerals crystallized simultaneously, and accumulated as they precipitated, with little or no sorting. In view of the much slower settling rate of Cr-spinel relative to olivine (Irvine and Smith, 1967) this suggests that settling distances were small (in the order of a few metres, or else spinel would "lag behind" olivine to produce cumulates barren with respect to spinel).

Assuming that olivine and Cr-spinel crystallized simultaneously and fractionation conditions were conducive to unimpeded gravitative segregation, and in view of the low spinel to olivine cotectic crystallization ratio and relative settling rates of the two minerals, the necessarily long accumulation times may severely limit concentration

of Cr-spinel by gravitative sorting mechanisms alone. Once olivine has begun to form, fractional crystallization by itself cannot yield a concentrated layer and some additional process is needed.

Irvine (1974a and 1977) has presented two alternative origins for chromitite based on phase diagram models of liquidus relations of Cr-spinel and olivine (fig 2.36). Both models place considerable significance on the SiO_2-rich spinel inclusions, and involve an increase in SiO_2-content of a liquid lying on the olivine-Cr-spinel cotectic. A change in the crystallization path is such that it crosses the Cr-spinel field,and therefore spinel only crystallizes. The 1977 model (fig 2.36) which involves mixing of primitive and differentiated liquids,is credible since multiple replenishment of magma chambers by new, generally hotter, primitive magma is well-evidenced from detailed petrological and geochemical studies of layered rocks (Brown, 1956; Irvine and Smith, 1967; Wager and Brown, 1968; O'Hara, 1977, Smewing, 1980), and evidently a process of major importance.

The model does not rule out the role of salic contamination, e.g. melted granitic country rocks (Irvine's 1974a model) or SiO_2-rich chamber floor (Huppert and Sparks, 1980, p. 286). However, in the case of Rhum, the greater abundance of inclusions in those chromitites within the olivine cumulates (than those overlying or underlying allivalite) may rule out the latter. To explain the sharp definition and lateral persistence of very thin spinel layers over >3 km strike distances, a new magma influx must have spread over the floor with great efficiency. If every chromitite layer and zone is the result of such a process, the implication is that the visible E.L.S. involved a considerably larger number of magma inputs than the fifteen suggested by the cyclic units (Brown, 1956). This point is discussed further in section 2.10. In the case of the chromitite "zones", sorting mechanisms

were such that several thin Cr-spinel layers formed instead of a single one, i.e. each zone is regarded as equivalent to a single magma mixing episode.

2.7.3.6 Sulphides: Due to limited sampling, sulphides have not been studied within the Central Series. However, sulphides occur in minor quantities (<0.5%) throughout most of the E.L.S. and above average concentrations (0.5-3%) are not uncommon. Concentrations are of disseminated-type only: matrix- and massive-types (10-60% and 60-100% sulphide respectively) are not recorded (terminology, Page, 1979). Sulphides are not homogeneously distributed throughout the sequence, but are associated with the Cr-spinel concentrations and the immediately overlying and underlying cumulates.

Several textural relationships are found between the sulphides and silicates. The former occur as (i) small rounded blebs (globules) within cumulus spinel and occasional olivine and plagioclase grains, (ii) subhedral smooth-edged interstitial patches adhering to cumulus silicates with no sign of replacement of the latter, and (iii) irregular angular straight-edged patches in the interstices between the silicates. The sulphide abundances are too low to produce continuous interconnection in 3-D.

On the basis of these observations, the sulphide minerals are regarded as comagmatic with the silicate material, having been precipitated as an immiscible sulphide liquid from the silicate parent liquid. The process of concentration is a gravitational one in which the sulphide liquid forms droplets which accumulate along with the silicates and oxides. Lack of massive sulphide layers is probably due to the limited volume of sulphide liquid produced, and the small settling distances involved - probably in the order of a few metres (larger distances would have resulted in the dense sulphides settling faster

than olivine and spinel, to produce density and modal grading).

Cu-Fe and Fe-sulphides are dominant (chalcopyrite, cp; bornite, bn; pyrrhotite, po; and cubanite, cb) followed by Fe-Ni-sulphide (pentlandite, pn) and minor amounts of Cu-sulphides (chalcocite, cc; and covellite, cv). Local alteration has produced small amounts of violarite, mackinawite, and pyrite. Each sulphide can occur individually or,more usually, in composite blebs with straight, gently-curving or highly-irregular mutual boundaries. The common assemblages are (i) cp, (ii) cp, bn, (iii) cp, bn, pn, (iv) cp, bn, cv, (v) cp, po, pn, and (vi) po, pn (fig 2.37). Similar disseminated Ni-poor assemblages have been described from the Skaergaard (Wager *et al.*, 1957), Muskox (Smith, 1962), Bushveld (Liebenberg, 1970) and Stillwater (Page, 1979) intrusions, Kilauean lavas (Skinner and Peck, 1969) and mid-ocean ridge basalts (Mathez, 1976).

No consistent correlations of sulphides are found with lithology, mineralogy, texture and stratigraphic position of the host rocks.

The mechanism(s) for separation of immiscible sulphide liquid from basaltic liquid require changes in physical or compositional parameters of part or all of the magma system. Such changes must enrich the system in sulphide liquid either by crystallization of other phases or by expanding the region of liquid immiscibility in the system. Information on the solubility of sulphide in silicate liquid, and the determining factors is limited. Skinner and Peck (1969) reported sulphur-saturation of Kilauea tholeiites at 0.038 wt% S and 1065^{o}C. Naldrett (1973, p. 9) quoted 0.16 wt% S for saturation of picrite at 1450^{o}C. Experimental studies suggest that the maximum amount of sulphur soluble in a basaltic magma varies between 0.05 and 0.2 wt% (Haughton *et al.*, 1974).

Activity of FeO in magmas is probably the main factor controlling sulphide solubility (MacLean, 1969) - the higher the activity of FeO,

the higher the sulphide solubility, and the less sulphide liquid is generated. Experimental studies (Naldrett, 1969; MacLean, 1969; Haughton *et al.*, 1974) suggest that the region of immiscibility is extended by (i) increasing sulphur fugacity f_{S_2}, (ii) increasing f_{O_2}, (iii) increasing SiO_2, alkali , MgO and Al_2O_3-contents in the magma, (iv) decreasing FeO-content, (v) decreasing total pressure, and (vi) decreasing temperature. The FeO-FeS-SiO_2 join in the system FeO-Fe_2O_3-FeS-SiO_2 is therefore of particular interest and is discussed below.

(i) Addition of sulphur has been suggested as a process, by Kullerud (1963), but there is no positive evidence that such a process occurred in Rhum: there is no known major source of sulphur in the country rocks.

(ii) If a process of rapid oxidation by e.g. incorporation of H_2O from country rocks is applicable, the oxide and silicate assemblages should show some evidence for high f_{O_2}'s (unless they re-equilibrated when the system became closed to the influx of H_2O and adjusted to the lower f_{O_2}). Evidence for high initial f_{O_2}'s and large volumes of water is scarce in the E.L.S. Some olivines show high-temperature oxidation in the form of dendritic exsolution of opaque oxide (Putnis, 1979), but the presence of exsolved ilmenite and metastable spinel in the Cr-spinels (Putnis and Price, 1979) suggests that oxidation occurred over a range of temperatures during subsolidus cooling (>600°C for the ilmenite and at a lower temperature for the intermediate phase). Intercumulus hydrous phases are absent in the cumulates, but the occurrence of biotite together with the sulphide, within the inclusions in the chromitite spinels, suggests that at the time of Cr-spinel (and therefore sulphide) precipitation, a significant amount of water was present.

(iii) and (iv) Changes in liquid composition (as specified above) may result from fractional crystallization or contamination. In the Rhum sulphide environments (within Cr-spinel layers) removal of FeO from the silicate liquid by the crystallization of large quantities of Cr-spinel may have been sufficient to initiate production of small volumes of sulphides. Alternatively a sudden increase in SiO_2-content of the silicate liquid may have moved it into the two-liquid field (sulphide and silicate, fig 2.38). The association of sulphides with chromitites strongly suggests that they were concentrated by the same processes as Cr-spinel (the mixing of new primitive picritic liquid with residual magma in the chamber - section 2.10.5).

The sulphide liquid which separated was Cu- and Fe-rich and clearly low in Ni, and therefore abundant nickeliferous sulphides did not develop in the E.L.S. The E.L.S. sulphides are low in Ni due to reduction in the Ni-content of the silicate liquid as a result of separation of Fe-Ni sulphides at an earlier unseen stage (evidence by the low Ni-contents of the E.L.S. olivines, section 2.9.1).

2.7.3.7 Ilmenite: Primary ilmenite is a minor interstitial phase only (<2 modal %), restricted to the plag-ol orthocumulates (allivalites of Units 6, 7, 8, 10) of the E.L.S., and rare occurrences in the olivine cumulates of both the E.L.S. and C.S.. In Units 6 and 7 it is the sole opaque oxide present.

Individual grains are anhedral, occur in the interstices between cumulus plagioclase or occasionally moulded onto olivine, and may compose a polycrystalline mosaic or a single crystal. They may be homogeneous or exsolve numerous small blebs, flames and laths of rutile (the R_2 stage of oxidation, Haggerty, 1976 page Hg 29).

Secondary ilmenite is ubiquitous throughout the L.U.C., exsolving from Ti-bearing Cr-spinel as a result of oxidation at moderate temperatures (>600°C) during subsolidus cooling. Trellis, sandwich and

composite types (analagous to those in oxidized titanomagnetite, Chapter 3, section 3.5.4.1) are found (fig 2.35). Eales and Snowdon (1979) have reported identical intergrowths from disseminated Cr-spinels within the slowly-cooled olivine cumulates of the Elephant's Head Dyke, and attributed them to an increase in f_{O_2}, and progressively retarded cooling rates.

2.7.3.8 Cumulate types: The textures now seen in the Rhum ultrabasic rocks are the end-product of a long history of cooling. Many, especially those in the plag-ol adcumulates, closely resemble the "metamorphic" textures seen in ultrabasic granulites and nodules (Vernon, 1970), and their interpretation must therefore take into account the probable solid-state readjustment of grain-shapes during slow-cooling in addition to simple mutual intergrowth of crystals (Hess, 1960).

In a purely descriptive sense, the terms ad, ortho and meso (Wager *et al.*, 1960) refer to cumulates in which (a) the cumulus phase(s) show unzoned, zoned, and both zoned and unzoned postcumulus overgrowth respectively, and (b) the intercumulus material (consisting of one or more phases other than the cumulus minerals) is inconspicuous or absent, abundant, and in minor amounts respectively.

However, in many layered intrusions, extreme ortho- and adcumulate textures may not be as common as once thought, due to the common difficulty in obtaining accurate values for the relative proportions of unzoned and zoned overgrowth. Many orthocumulates may have had some initial adcumulus growth.

Several important points regarding the distribution and relative abundance of various cumulate types in the Rhum intrusion are:
(i) the E.L.S. and C.S. peridotites (both harrisitic and non-harrisitic) are heteradcumulates, i.e. cumulus olivine is unzoned and the rocks

contain moderate abundances (section 2.7.1) of large virtually-unzoned (section 2.9) poikilitic intercumulus clinopyroxene and plagioclase plates.

(ii) the relatively olivine-rich plag-ol (±cpx) cumulates in both E.L.S. and C.S., are mesoadcumulates, containing again unzoned olivine, cumulus feldspar (and clinopyroxene) with narrow often strongly-zoned overgrowths, and minor (<5%) amounts of intercumulus pyroxene (non-poikilitic).

(iii) orthocumulate textures are confined to the olivine-poor plag-ol and cpx-plag-ol cumulates; the former in the uppermost parts of Units 6 and 11 and the basal portions of Unit 8 and 10 allivalites (fig 2.30), the latter in the Dornabac Member. Cumulus plagioclase is strongly zoned (cumulus olivine is not), and intercumulus phases include clino-pyroxene, ilmenite ± kaersutite.

(iv) extreme olivine adcumulates (dunite) are absent from the E.L.S., but are common in the Long Loch and Ruinsival Members.

(v) extreme plagioclase adcumulates (anorthosite) are very limited, being restricted to the topmost 0.5-10 cm immediately underlying chromitite layers (E.L.S. only).

(vi) there is no discernible systematic lateral variation, along-strike or down-dip, in major cumulate types of the E.L.S.. In contrast the Central Series does show considerable along-strike variation, e.g. in the Long Loch Member.

(vii) olivine supersaturation textures (harrisite and comb-layered harrisite) are abundant in the E.L.S. peridotites, and virtually absent in the E.L.S..

In view of the long crystallization history of these rocks, their textural interpretation, especially regarding the genetic significance of the degree of mineral zoning, must be treated with caution (see also

section 2.10). Several processes which may have operated within the cumulate pile to modify, or destroy, the original textures and mineral compositions are:

(a) continuous reaction with migrating intercumulus liquids (infiltration metasomatism, Irvine, 1980b). This was probably limited to the early stages of solidification, when compaction and postcumulus overgrowth were insufficient to impede the free movement of liquid through the interstices. Initial porosity of crystals in accumulating mush is estimated at between 20 and 50% (Jackson, 1961; Wager and Brown, 1968).

(b) reaction with trapped intercumulus liquid (when "permeability" had been significantly reduced).

(c) continuous solid-state readjustment and re-equilibration (possibly with recrystallization) during slow-cooling, which would act to homogenize grains.

Several workers (Wager, 1963; Campbell, 1968; Goode, 1977) have postulated that production of adcumulate textures requires intercumulus environments "open" with respect to the overlying liquid, slow accumulation rates (for effective diffusional exchange), and relatively high rates of intercumulus crystallization, i.e. the liquid is supercooled and supersaturated with respect to the cumulus phases. Such conditions it was believed could only be found at or near the top of the cumulate pile. If accumulation was fast, much of the intercumulus liquid was trapped and orthocumulate textures resulted.

However, Irvine (1980b), as a result of his infiltration metasomatism modelling, supported Hess' (1972) conclusion that adcumulus growth does not only occur at the cumulate depositional surface. He presented relatively direct evidence that a limited amount of adcumulus growth can occur deep in a thick, unconsolidated cumulate pile

(often hundreds of metres below the depositional surface) as a result of compaction, and the local removal of "unwanted" components by diffusion in upward-migrating intercumulus liquid or by diffusion through stationary resident liquid (in response to thermal and compositional gradients ahead of an upward-advancing solidification front). The process is still not fully understood or explained, and requires detailed investigation of (a) relative heat and mass transfer rates within unconsolidated crystal piles, (b) relative diffusivities of the various cations involved, (c) the changing values of porosity and permeability with time, (d) the possibility that convection could occur within the intercumulus liquid such that less fractionated liquid moves down to replace upward-filtering liquid (i.e. a process of double-diffusive fractional crystallization, Irvine, 1980b). All are beyond the scope of this thesis.

2.7.3.9 Supersaturation textures: That a considerable proportion of the Rhum L.U.C. crystallized under conditions of supersaturation is suggested by such phenomena as the harrisitic olivine (Wadsworth, 1961; Donaldson, 1974, 1975, 1977a) and poikilo-macrospherulitic feldspar (Donaldson *et al.*, 1973) textures. Distributions of the two textures are markedly different. The supersaturation olivine morphologies are abundant in the E.L.S. and extremely rare in the C.S., whilst the supersaturation plagioclase morphologies (intercumulus only) are confined to the Central Series. Cumulus plagioclase and both cumulus and intercumulus clinopyroxene do not show corresponding textures. Similar elongate branching or curved supersaturation crystal morphologies are reported from a variety of other shallow-seated coarse-grained igneous intrusions, e.g. Skaergaard (Wager and Brown, 1967), Willow Lake (Taubeneck and Poldervaart, 1960), and from volcanic rocks (Bryan, 1972; Pyke *et al.*, 1973), and have been experimentally produced

(Donaldson, 1977a; Lofgren *et al.*, 1974; Lofgren, 1974).

The origin of the Rhum supersaturation textures is discussed with a view to elucidating their observed distribution within the various layered series in terms of differing crystallization conditions. As described in section 2.4, harrisitic olivines within the E.L.S. form numerous widespread (laterally persistent) sharply- and gradationally-defined layers, alternating with "normal" olivine cumulates. In contrast, only two harrisitic horizons have been recorded within the C.S. (section 2.5) - one thick zone immediately overlying the Outer Breccia Zone (both E and W of the Long Loch Fault) and one thin layer in Unit A of the Stable Flats area. Supersaturation feldspar textures are similarly confined to sharply-defined layers at essentially constant stratigraphic horizons over a strike distance of ca. 0.5 km (Central Series only), e.g. the poikilo-macrospherulitic facies NE of Loch an Dornabac (section 2.6.3 and Donaldson *et al.*, 1973), and at the margins of the Ruinsival Member peridotite on Barkeval.

Supersaturation in subvolcanic magma bodies is generally the result of one or more of the following mechanisms (i) conductive cooling at margins, i.e. heat loss into country rocks, (ii) cooling due to intrusion into a larger, cooler unit of magma, (iii) elevation of liquidus temperature relative to the actual temperature.

In the case of the Rhum intrusion, the olivine supersaturation morphologies occur well within the E.L.S., where cooling effects on the roof or sides might be expected to be minimal (Irvine, 1970b). However, cessation of convection, either throughout the chamber or in a unit at the floor, can induce supersaturation by altering the temperature gradient at the base of the chamber from one in which temperature increases towards the cumulate pile (0.3°C/km, Jackson,

1961) to one where temperature decreases towards the pile (Donaldson, 1975a), i.e. non-convecting magma at any given height is cooler than at the same height during convection. With this mechanism it is difficult to envisage convection cyclically ceasing and restarting so repeatedly as to produce the numerous, often <1 m, layers alternating with "normal" olivine cumulates.

Initiation of olivine supersaturation textures due to a change in the degree of liquid supercooling as a result of injection of fresh (hotter) magma or liquid into the chamber (mechanism (ii)) is in accord with Brown's (1956) idea of replenishment, but the process was rejected by Donaldson (1975a and 1977a) on the grounds that (a) it would require cyclic inputs on a scale of ca. 1 m, (b) harrisite is not found between units in the E.L.S., (c) it would be unlikely for a fresh input of material to produce a perfect continuous layer often <1 m thick, (d) there is no textural or structural evidence for intrusional movement of liquid prior to harrisite formation, and (e) experiments suggest continuous, rather than abrupt, changes are likely in the pressure, temperature and other parameters of the harrisite-forming liquid.

However, in view of data presented in sections 2.9.4, 2.9.6, 2.10 and Chapter 6, mechanism (ii) need not be immediately rejected on the grounds of the five objections mentioned above; e.g. the number of fresh influxes into the chamber may be greater than the 15 suggested by the cyclic units (Brown, 1956), and the parental material is regarded as being a high-MgO (>9%) basalt (Chapter 6). Huppert and Sparks' (1980) model for the spreading of layers (of the order of a few metres) of hotter more primitive (higher density, lower viscosity) liquid over the base of a chamber containing significantly cooler, less dense, basaltic liquid (<9 wt% MgO), predicts that thin layers are likely to

show a combination of rapid cooling rates and relatively low degrees of turbulence (op. cit. figs 5 and 6), i.e. conditions of a sufficiently high degree of supercooling to initiate crystallization of skeletal olivine morphologies. The paucity of nucleation sites per unit volume, and extreme grain-size of the olivines are most likely the result of an antipathetic relationship between nucleation and growth rates, such that when the nucleation rate was low, the growth rate was near to the maximum for olivine at the considerable degrees of supercooling. Convincing evidence for the formation of quench textures in layered igneous intrusions during influx of hot, primitive magma into a basaltic chamber has been reported from the Hettasch Intrusion, Labrador (Berg, 1980), where plagioclase displays spectacular comb layering and large-scale spherulitic textures.

Application of the Huppert and Sparks (1980) model of harrisite formation to the Rhum E.L.S. (and W.L.S.) would seem to imply that each harrisitic (and protoharrisitic) layer formed from a single, thin, rapidly-cooled, poorly-convecting layer of primitive liquid, by the crystallization and settling of skeletal olivine crystals. If this is true it is difficult to envisage how successive batches of liquid (of the same composition and temperature) can suffer differing degrees of supercooling or how some layers are rapidly cooled (to give harrisite) and others not (normal cumulates). Alternatively, the model can be modified to emphasize the gradational nature of the contacts between harrisitic and non-harrisitic layers, stress the genetic relationship between the two, and do away with the need for repeated large changes in the degree of liquid supercooling. Donaldson (1977a) suggested that if a slightly-supercooled magma becomes sufficiently cleared of crystals, at a rate greater than the nucleation rate (i.e. the nucleation rate is effectively decreased), supersaturation locally increases

(less competition between crystals for solute), therefore growth rate increases and skeletal morphologies are produced. This suggestion may be consistent with Huppert and Sparks' (1980) hypothesis, in that if conditions were such that the olivine settled en masse (as the two layers approached thermal equilibrium and convection died down in the lower layer) then this rapid loss of crystals may have been sufficient to promote crystallization of supersaturated morphologies. The low viscosity of the picritic parental liquid would aid this process by promoting rapid sinking of the crystals.

An alternative origin is required for the occasional harrisite layers which show vertical alignment of the olivine crystals (comb-layers), and require conditions of (a) weak convection i.e. little disturbance of the thermal gradient, (b) upward flow of latent heat of crystallization into supercooled liquid, (c) low concentration of suspended crystals, and (d) a substrate (Donaldson, 1977a). Irvine (1980b) envisaged comb-layers forming where the bottom contact of the lowest convection layer is essentially coincident with the top of the cumulate pile, and is the junction between the main liquid body and the intercumulus liquid filtering from below (Irvine, 1980b, fig 20). The olivines grow in the diffusive interface, and constituents for their growth are drawn from the overlying liquid which has been super-cooled by the convection process.

Elevation of olivine liquidus temperature as a mechanism for the promotion of supercooling, may result from changes in confining pressure, f_{O_2}, and H_2O-content of the liquid or magma. A decrease in confining pressure implies a fresh magma pulse per harrisite layer, which as mentioned above may not be feasible, whilst large decreases in oxygen fugacity (by ca. six orders of magnitude) would induce supercooling of only 12°C (Biggar, 1974), and would decrease Fo-content of the olivine

(which is not found). A mechanism involving diffusional loss of water along a pressure, temperature or compositional gradient, either through a uniformly water-undersaturated liquid column or within a stratified column, has been tentatively suggested as the cause of supersaturation (Donaldson, 1975a). Although the water-rich nature of the picritic parental liquid is indicated by the abundance of picritic dykes rich in hydrous phases (Chapter 6) it is questionable whether the elevation of liquidus temperature by water diffusion is fast enough to induce supersaturation (Donaldson, 1975a).

Considering all the arguments presented above, it is tentatively suggested that the bulk of the field, petrographic and mineralogical data supports induction of supersaturation by replenishment of fresh (hotter, more basic) magma onto the floor of the chamber. Depending on (a) the thickness of the newly-introduced layer, (b) its temperature relative to the resident liquid in the chamber and the intercumulus liquid in equilibrium with the crystals on the floor, and (c) the style and relative roles of bottom crystallization, and convection, the relative abundance of normal to harrisitic cumulates produced from the introduced layer will vary from influx to influx. If alternating periods of large-scale convective turnover and stagnation were the primary cause of supersaturation (Donaldson, 1975a) then similar conditions and harrisite cumulates should be widespread in other sizeable magma chambers. That they are not, implies clearly that there was something almost unique about the solidification of the Rhum parent liquid which promoted harrisite crystallization. Data from this and Chapter 6 suggest the likely factors are the water-rich and MgO-rich nature and the repetitious influx of hot primitive magma.

Rarity of olivine supersaturation textures in the Central Series is a direct consequence of intrusion of magma batches already containing

a large proportion of olivine crystals in suspension (section 2.6). Plagioclase supersaturation textures developed in the intercumulus liquid instead. Volumetrically they are less important in the C.S. than harrisite is in the E.L.S., and do not alternate with normal cumulates in a regular fashion, and it is therefore implied that either the number of replenishment episodes was smaller, or, as is more likely, the supercooling was induced by a different, less frequently operating mechanism. For example, the occurrence of poikilo-macrospherulitic texture in the marginal zone (a few metres wide) of the Ruinsival Member peridotite suggests induction of supersaturation by conductive heat loss into adjacent rocks (these adjacent rocks - gabbros and dykes - show evidence of thermal metamorphism - Chapters 3 and 4). An additional contributing factor may have been a rapid reduction in water pressure (the hydrous nature of the intrusive magma is indicated by the abundance of biotite and amphibole, relative to the E.L.S.). This water loss may have occurred during rapid uprise of the Central Series porphyritic magma, or subsequent to emplacement e.g. during eruption of an associated volcano. It is dependent on (i) the difference in water pressure between magma and country rocks, (ii) rate of flow of water into the country rocks, (iii) solubility of water in the magma (which decreases with decreasing pressure for water-undersaturated systems), and (iv) the temperature difference between country rocks and magma. As in the case of the skeletal olivines, the paucity of nucleation sites per unit volume, and consequently the large grain-size of the feldspars involved in formation of this texture, are the result of a high degree of supercooling and simultaneous high growth rate (Lofgren, 1974).

2.8 Origin of the Igneous Layering

2.8.1 Macro-layering

In the widely-used classification of igneous layering introduced by Wager and Deer (1939), a principal type is "rhythmic layering", in which individual layers are a few cm to 1 m thick, and defined by rhythmically repeated conspicuous differences in cumulus mineral assemblage. On a large scale (tens or hundreds of metres) such variation has been termed "macro-rhythmic" layering, and is a prominent feature of many layered basic and ultrabasic intrusions. When the layered sequences are composed of a number of large, regular, repetitious units the term "cyclic" is used. The Rhum units more closely resemble the cyclic units of the Stillwater and Muskox intrusions (Jackson, 1961; Irvine and Smith, 1965) and are referred to by that name, as in a recent paper by Dunham and Wadsworth (1978).

Although macro-layering is well-documented, its origin is still uncertain, and has been explained by three contrasting theories: (i) multiple intrusion (Brown, 1956; Irvine and Smith, 1967; Irvine, 1974; Campbell, 1979), (ii) alternating depletion and replenishment of a basal zone of stagnant liquid, by periodic convection (Jackson, 1961), and (iii) rhythmic nucleation events in the magma (van Zyl, 1959; Wager, 1959; Hawkes, 1967; Maaloe, 1978; Goode, 1977). Other proposed mechanisms involve fluctuation of physio-chemical conditions or liquid composition as a result of events of external origin, e.g. changes in f_{O_2} and pressure (Cameron, 1969) and loss of volatile components (Yoder, 1955). Although these latter processes may be important, it remains to be proven that they are cyclical. Considering the similar layering within different intrusions, and the varying styles of layering within a single intrusion, it is unlikely that each intrusive body had a similar mechanism.

Jackson (1961) justified the introduction of a variable-depth convection model by arguing that the theory of multiple injection encounters several difficulties when applied to Stillwater, including (a) the absence in the lower units of cross-cutting feeder dykes to the higher units, and (b) the absence, at the base of each macro-unit, of Fe-Ni-Cu sulphide concentrations such as are at the base of the intrusion (i.e. from the initial injection). His model involves crystallization at, or near, the base of the chamber from a layer of liquid which is supercooled as a result of heat loss into the cumulate pile. Crystallization within this layer raises its density and prevents it convecting, although the remainder of the liquid in the chamber is free to convect. When latent heat of crystallization raises the temperature of the stagnant layer above the liquidus temperature, further crystallization is impossible and the layer joins in the convection system. The cycle restarts when a major convective overturn sweeps away the stagnant layer and replaces it with more primitive liquid. This mechanism operates without infusions of new magma and is thought to be self-damping.

Jackson's first objection to the multiple intrusion hypothesis ((a) above) may not apply to Rhum, since the Long Loch Fault region, which acted as a feeder to the Central Series (Chapter 7), may also have fed the Eastern Layered Series. Objection (b) also does not apply, since above-average concentrations of sulphides occur at the base of many units (section 2.7.3.6). Jackson's variable-depth convection model, involving only one major magma injection, accounts for the progressive increase in amounts of lower-temperature phases in the higher cyclic units of the Stillwater complex. Since such a feature is not found in Rhum (section 2.10) Jackson's hypothesis (1961) is not considered further.

In contrast, convincing evidence for the repeated influx of new (more primitive) magma into a chamber is provided by geophysical investigations of active volcanoes (e.g. Kilauea, Krafla), and the detailed petrological and geochemical studies of igneous rocks from many different volcanic environments: mid-ocean ridges (O'Hara, 1977; Donaldson and Brown, 1977; Walker *et al.*, 1979), island arcs (Anderson, 1976) and continental areas (Smith, 1979). It has been convincingly demonstrated in many basic and ultrabasic intrusions that macro-layering marked by major reversals of crystallization trends can only be explained by multiple injections of fresh magma (Rhum: Brown, 1956; Muskox: Irvine and Smith, 1967, Irvine, 1979; Jimberlana: Campbell, 1977; Oman ophiolite: Smewing, 1980). Magma replenishment generally prolongs the life of a chamber by providing additional thermal energy (Usselman and Hodge, 1978) and a mechanism for eruption at the surface (Sparks *et al.*, 1980), and is obviously a process of great petrological importance.

Macro-layers are internally coherent as to textures, grain-size etc., but are stratigraphically sharply-defined. The cumulus succession within any given cyclic unit is that predicted by the liquidus paths determined experimentally on simplified basalt systems (Jackson, 1970). The cyclic units are therefore thought to represent simple fractionation of small batches of magma, terminated by horizons representing introduction of a new magma batch and repetition of the crystallization sequence.

There are three main models for bringing fresh influxes of magma into a chamber (Campbell, 1977; Huppert and Sparks, 1980):

In model 1, injection of a sudden pulse is followed by rapid homogenization with the resident magma, thus modifying the composition of the latter and raising its temperature. During the period of

adjustment the temperature at the top of the cumulate pile rises, and gives rise to reversed mineral variation (assuming Jackson's (1961) concept of bottom crystallization).

In model 2, slower injection is extended over a period of time, represented by the zone of mineral composition reversal.

In model 3, the primitive magma injected does not immediately mix with the resident (more evolved) magma, but spreads over the floor forming an independently crystallizing layer (Huppert and Sparks, 1980). Only when the density of this layer is sufficiently reduced (by olivine crystallization) does it become buoyant and able to mix with the resident magma.

These three models are evaluated in section 2.10.5 using compositional variation data for both cumulus and intercumulus phases.

2.8.2 Fine-scale layering

Within the E.L.S. small-scale (<0.5 m) variations in cumulate mineralogy and textures are typically superimposed on the macro-scale rhythmic layering. Layers within the olivine (+ spinel) cumulates are defined by abrupt changes in olivine grain-size and/or morphology, and are not internally graded. Layers in the plag-ol-sp cumulates are commonly modally-graded, involving abrupt alternating variations in modal proportions of the three main cumulus phases, but little systematic differences in grain-sizes.

Until the 1970's the field and textural data on a large number of ultramafic and gabbroic intrusions had been interpreted satisfactorily in terms of gravitational accumulation of crystals (Wager and Brown, 1968; Irvine, 1971). Descriptive sections noted the similarity of rhythmic layers to graded-bedded turbidity current deposits, and suggested that they were probably deposited in an analagous fashion by magma currents. Some idea of the mechanisms involved was given by

flume experiments on turbidity currents (especially those by Middleton, 1967). Irvine (1980a) extended Middleton's experiments with a view to examining in detail the internal structures of magmatic density currents. He described possible mechanisms whereby currents may deposit graded layers, and suggested mechanisms for the production of thinly-layered or uniform olivine cumulates by current deposition and bottom crystallization. Data from the Rhum intrusion appear to support his models. Because the modal-graded layer consists of crystals deposited by the top and bottom portions of the suspension current (Irvine, 1980a, fig 10) it might be predicted that natural graded layers would be vertically bimodal in mode and/or grain-size. In the E.L.S. olivine cumulates the better-sorted layers (which are relatively thin and numerically, though not volumetrically, significant within the total sequence) are indeed characteristically bimodal in respect of grain-size. Typically they exhibit a rapid transition from a relatively coarse peridotite at the base to a distinctly finer peridotite at the top, often with the effect of appearing as two layers (section 2.4.2).

Discussion of layering in the allivalites raises the question of whether the plagioclase crystals were capable of gravitative settling. For intrusions in which the liquid became very iron-rich (Skaergaard, Kiglapait, Bushveld) workers have estimated that the plagioclase was less dense than the liquid from which it crystallized and so, if anything, should have floated (Wager and Deer, 1939; Bottinga and Weill, 1970; McBirney and Noyes, 1979; Campbell *et al.*, 1978; Morse, 1979; Willemse, 1969). However, since the Rhum liquid never reached such iron-rich compositions (as indicated by mafic mineral compositions) the plagioclase-flotation problem probably does not arise. Probably a more important factor relates to the nature of the magma itself. It is now well-established that crystallizing basic magmas are

non-Newtonian fluids with substantial yield strengths which must be overcome before a crystal will move (McBirney and Noyes, 1979). Plagioclase's density contrast with basaltic liquid is generally so small (Bottinga and Weill, 1970) that its distribution can be expected to be very sensitive to convective movements.

Returning to the question of the origin of the fine-scale layering within the allivalites, the role of current deposition is considered. The layering features thin discontinuous layers consisting of alternate laminae of predominantly mafic phases and plagioclase, with periodically interspersed groups of thicker modally-graded layers. Because of their frequent and discontinuous nature, it is unlikely that each lamina (<5 cm thick) could be explained by a separate density current, but they could possibly be explained in terms of repeated separation from continuous currents (produced by convective overturn); with only episodic rapidly-moving density currents producing the graded layers (Irvine, 1980a, fig 11). Features which may indicate current transport and deposition include anorthosite fragments (a few cm in length) abundant in many allivalites, and which often show a distinct imbrication (first recorded by D. Godfrey). These "fragmental layers" (Irvine, 1980d) are well-defined and regular in thickness, implying that the disturbed matrix around the fragments was carried along with them from some other part of the intrusion, before being deposited upon the floor (cf. Units B and C of the Long Loch Member, section 2.6.4). Analagous fragmental layers and associated disturbed zones have not been identified with the olivine cumulates.

Although gravitational deposition from moving currents may be a major process involved in the formation of the layering styles characteristic of the allivalites and peridotites, the role of in situ crystallization (with or without crystal settling) and double-diffusive convection cannot be ruled out.

A mechanism of in situ bottom crystallization was proposed by Jackson (1961), based on the observation that the liquidus temperature of magma increases more rapidly with depth (ca. $3^{\circ}C/km$) than the anticipated adiabatic gradient in a convecting magma, but as pointed out by Irvine (1980a) one problem with this hypothesis is that, in the case of cumulus layers forming a few 100 m below the roof, crystallization is implied to have occurred at the floor rather than the roof because of a temperature difference of $<1^{\circ}C$. However, this problem is overcome if, as has been suggested (sections 2.9.1 and 2.10), repeated influxes of hotter, more primitive liquid were ponded across the floor as the result of a density difference with the resident magma. In such a situation the increase of liquidus temperature with depth would be a function of composition as well as of pressure.

Indications are that, despite the importance of magmatic current transport and deposition of crystals, the majority of crystals in layered rocks have formed by in situ crystallization (Irvine, 1980a, b). Theoretical and experimental investigations have revealed that even very small compositional differences between resident and introduced magmas can give rise to density stratification, and that such stratification can exert a dominant control on convection systems and crystallization processes. The term "double-diffusive convection" has been used (McBirney and Noyes, 1979), since the process involves heat and mass transfer through coupled diffusion and convection. A detailed description of the process is beyond the scope of this section, and the reader is referred to detailed reviews by Irvine (1980b, c). This new concept offers numerous possibilities for the reinterpretation of layering structures previously ascribed to crystal settling. However, it requires rigorous testing by experimental, and

more especially field, investigations. For example, McBirney and Noyes (1979) have advocated that double-diffusive convection could produce graded and other kinds of modally differentiated layers, but do not suggest a specific mechanism, and Irvine (1980a, b) envisaged the formation of vertically-aligned crystals (e.g. comb-layered harrisite) in circumstances whereby the base of the lowest convection layer is essentially coincident with the top of the cumulate pile, and the olivines grow in the diffusive interface between the main magma body and intercumulus liquid diffusing out of the cumulate pile.

Therefore, with regard to the question of gravitational segregation of suspended crystals from flowing magma currents versus in situ crystallization, it is tentatively suggested that modally-graded layers in the 2 or 3-phase cumulates (the allivalites) are gravitationally-produced current deposits, and that if in situ crystallization was also important in the E.L.S. it is more likely to have occurred within the olivine cumulates.

The lack of fine-scale rhythmic layering within the olivine cumulates of the Central Series, and the scarcity of evidence for magmatic currents and/or in situ crystallization, are directly related to the nature of the intruded material. Field evidence (section 2.6) suggests that in contrast to the aphyric (<10% crystals) magma entering the chamber during formation of the E.L.S., the C.S. formed from highly olivine-phyric magma (>50% crystals), for which the term "crystal-mush" could alternatively be employed.

2.9 Mineral Chemistry

Mineral compositions were determined by microprobe analysis, using the method described in Appendix A. Precision and accuracy values are presented in Table A.2. Stratigraphic (cryptic) variations in mineral compositions are described and discussed in section 2.10.

2.9.1 Olivine

Olivine is a cumulus phase throughout the L.U.C., ranging $Fo_{89.4}$-$Fo_{79.1}$ and $Fo_{87.1}$-$Fo_{78.1}$ in the olivine and plagioclase-olivine cumulates respectively. The overall variation is identical in both E.L.S. and C.S. rocks (Fo_{89}-Fo_{78}). Comparable ranges are shown by the Great Dyke (Fo_{94}-Fo_{84} Worst, 1958), Stillwater (Fo_{94}-Fo_{80} Jackson, 1961), Muskox (Fo_{93}-Fo_{83} Smith and Kapp, 1962; Irvine, 1970), and Duke Island (Fo_{91}-Fo_{78} Irvine, 1974b) complexes.

The apparent range of compositions in each rock sample is small (maximum 1.0 mol % Fo) and within the limits of analytical error. Zoning is absent.

CaO-contents of the olivines (fig 2.39) range from 0.16-0.02 wt % (for both peridotites and allivalites) and show little change with fractionation. These uniformly low contents lie within Simkin and Smith's (1970) range for "plutonic" olivine (<0.2%) and are comparable to those reported from alpine-type peridotites, peridotite nodules and other layered intrusions (fig 2.39). That they are lower than the levels predicted (0.16-0.30 wt%) for olivine in equilibrium with a basaltic liquid (av. 6-11 wt% CaO, Watson, 1979) suggests Ca has been lost to the surrounding silicates during slow-cooling and subsolidus re-equilibration (Ferguson, 1978). Such diffusional loss of Ca may explain the lack of expected lower CaO-levels in olivines which have crystallized with plagioclase.

MnO-contents (fig 2.40) increase linearly with fractionation from 0.17 wt% at Fo_{89} to 0.33 wt% at Fo_{79}, for both olivine and plag-ol cumulates. Within-sample variation is small (av. 0.02 wt%).

NiO-contents are highly variable (fig 2.41) and the two layered series (E.L.S. and C.S.) show different NiO-Fo patterns. Central Series olivines are richer in NiO than those in the E.L.S., and show a roughly

linear (albeit scattered) decrease with fractionation from 0.40 wt% at Fo_{89} to 0.22 wt% at Fo_{79}. In contrast, the E.L.S. olivines show a less ordered variation. Those in the peridotites have a wide range of NiO-values over a restricted Fo-range (0.37-0.14 wt% for Fo_{86-84}), whilst in the allivalites, NiO (0.37-0.12 wt%) shows a poorly-defined linear decrease with increasing Fa-content. Factors which may have produced the observed Fo-NiO patterns include (a) loss of Ni during alteration, (b) decreasing olivine-liquid Ni distribution coefficient (D_{Ni}^{ol-liq}), and (c) crystallization of olivine from Ni-depleted liquids.

Although serpentinization can deplete olivine in Ni (Coleman and Keith, 1971), its lack of correlation with NiO-content precludes the importance of this process in the case of Rhum.

D_{Ni}^{ol-liq} apparently varies with temperature (Leeman, 1973; Leeman and Lindstrom, 1978), pressure (Mysen and Kushiro, 1979) and Ni-content of the liquid (Arndt, 1977; Hart and Davis, 1978; Elthon and Ridley, 1979). NiO-poor olivines, in the case of Rhum, cannot be ascribed to increasing pressure (which decreases D_{Ni}^{ol-liq}) since the liquids are assumed to have been retained within a single high-level chamber. The dominant controlling factor for lowering D_{Ni}^{ol-liq} is believed to be a decreasing Ni-concentration in the liquid (Clarke and O'Hara, 1979; Elthon and Ridley, 1979), which would be most effectively achieved by the fractionation of Ni-bearing sulphides (with a very high $D_{Ni}^{sulph-liq}$ value, ca. 275, Rajamani and Naldrett, 1978). The observation that pentlandite is a common trace constituent in many of the Rhum cumulates (section 2.8.7) lends support to this hypothesis.

Duke and Naldrett (1978) and Duke (1979) have modelled the variation in NiO-contents of olivine which fractionated from sulphur-saturated (i.e. sulphide-precipitating) and sulphur-undersaturated (sulphide-free) mafic and ultramafic liquids: their results are

summarized in fig 2.41. Curves marking the maximum NiO-values for given Fo-contents of olivine, from Rhum and mantle-derived nodules are superimposed on the calculated data of Duke and Naldrett (1978). The discrepancy between the modelled sulphur-undersaturated curves and the natural data may be due, not to incorrect assumptions regarding the partitioning behaviour of Ni, but to unrealistically low Ni-concentrations in the initial liquid (1600 ppm Ni at 32 wt% MgO compared to 1900 ppm for similar MgO, reported by Nesbitt and Sun, 1976). However, the parallelism and geometric similarity of the calculated and natural data curves suggest that the overall conclusions regarding the differing NiO-contents of olivine crystallizing from sulphur-saturated and undersaturated magmas, are valid. Whilst accepting the possibility of modification of sulphur-saturated trends by fractionation involving varying ratios of olivine to sulphide (a decrease in this ratio, as a result of decreasing sulphur solubility with ongoing fractionation, Haughton *et al.*, 1974, leads to enhanced Ni-depletion trends, Duke, 1979), the modelling of Duke and Naldrett (1978) involved realistic estimates of this ratio (in the order of 1000).

Several important petrogenetic conclusions can be made regarding the L.U.C. data in fig 2.41. Olivines from the Central Series lie on a curve parallel to, but non-coincident with, the Rhum maximum, and appear to have crystallized from a sulphur-undersaturated magma whose initial Ni-content was less than that for the Rhum parental liquid (on the "maximum NiO" curve).

In contrast, olivines from the E.L.S. have equilibrated with a sulphur-saturated magma. The Fo-NiO configuration for olivine in the peridotites suggests that successive batches of magma became sulphur-saturated (a) at approximately the same stage of differentiation (at Fo_{85}, i.e. ca. 12% MgO in the liquid, using olivine-groundmass pairs

in Table 6.5), and (b) prior to, or simultaneously with, initiation of olivine (Fo_{86}-Fo_{84}) fractionation (i.e. the case of curve 1 or 2 in fig 2.41). This latter conclusion implies that the liquid entering the E.L.S. chamber carried only a small proportion of olivine crystals (ca. 10%).

Olivine in the allivalites shows greater spread of NiO-contents (fig 2.41). The higher NiO-contents of olivine in Units 7 and 11 allivalites (relative to those in 6, 8 and 9) suggest that the liquid from which they grew became sulphur-saturated at a much later stage in its history (paralleling a less steep Fo-NiO curve, e.g. curve 3 in fig 2.41).

The relatively barren nature of the Rhum ultrabasic cumulates, with regard to sulphide mineralization, is at variance with the conclusions reached above. It is possible that the E.L.S. liquids (or magmas) may have separated a sulphide liquid at some stage before their entry into the magma chamber, which by some means, possibly gravitationally controlled, became separated from the parent silicate liquids. Ni-sulphide concentrations are therefore predicted at depth in Rhum. In the case of Units 11 and 12 which became sulphur-saturated at a much later time (i.e. within the magma chamber), the liquid retained its sulphide liquid: hence the above-average modal contents of sulphides in these rocks (ca. 4%). Differentiation conditions were such as to prevent the concentration of sulphide by gravitational processes: the sulphides are disseminated throughout the units.

2.9.2 Plagioclase

Plagioclase is postcumulus (poikilitic) in the peridotites, and cumulus in the allivalites (E.L.S.) and Dornabac and Long Loch (Unit B only) Members (C.S.). The range of compositions (fig 2.42) is

extensive and similar for both varieties of feldspar, from An_{92} to An_{53}. Cumulus plagioclase is generally more calcic in the E.L.S. (An_{90}-An_{81}) than in the C.S. (An_{83}-An_{79}), whilst poikilitic plagioclase is similar in both series; $An_{87.5}$-An_{77} (E.L.S.) and $An_{85.5}$-An_{74} (C.S.). Similar variations are found in the Salt Lick Creek (Wilkinson *et al.*, 1975), Lilloise (Brown, 1973) and Skye Cuillin (Hutchison and Bevan, 1977) intrusions.

Moderate to strong marginal zoning extends cumulus and intercumulus ranges to An_{64} and An_{53} respectively. However, in many intercumulus and some cumulus grains, zoning is reversed (the rims being more calcic than the cores by an av. of 4 mol %, and up to 10 mol % An). Similar profiles have been produced in plagioclase crystallized experimentally at a single temperature and relatively high degree of supercooling (Lofgren, 1974). Lack of associated supersaturation textures (e.g. poikilo-macrospherulitic) may be a function of the degree of packing of the olivines. The origin of supersaturation textures (and supercooling) has been discussed in section 2.7.4.9.

More pronounced within-sample variations, excluding obvious marginal zoning, are found in the intercumulus (maximum 6 mol % An) than in the cumulus grains (maximum 2 mol % An). This 6 mol % An range is surprising for rocks described by previous workers as adcumulates. For a more detailed discussion see section 2.10.

2.9.3 Clinopyroxene

As a cumulus phase, clinopyroxene is restricted in occurrence to the uppermost few metres of Units 8, 9 and 10, together with the Dornabac Member and localized horizons in the Long Loch and Ruinsival Members (M51 and B34), but it is present as a postcumulus phase throughout the complex. All pyroxene is free of pigeonite and orthopyroxene exsolution.

2.9.3.1 Major element variation: Analysed clinopyroxenes from both the E.L.S. and C.S., expressed in terms of atomic ratios Fe : Mg : Ca (where Fe = total iron as Fe^{2+}), have been plotted on the pyroxene quadrilateral (fig 2.43). Five features are noteworthy:

(i) compositional variation is very limited (diopside only): $Fe_{5.5-10}$ (majority 6-8) at constant Ca:Mg ratio, and Ca_{49-44} (majority 47-45) at constant Fe:Mg.

(ii) there are no compositional differences between pyroxenes (both cumulus and intercumulus) of the E.L.S. and C.S., and between cumulus and intercumulus types in general.

(iii) the few analysed intercumulus grains from orthocumulate allivalites (Units 6, 7, 8), tend to be poor in Ca (by 2-3 atomic %) relative to meso- and heteradcumulates (the reverse of variation reported from the Jimberlana intrusion, Campbell and Borley, 1974), due to growth from trapped liquid depleted in Ca by plagioclase overgrowth.

(iv) fractionation (marked by within-crystal variation) takes place by the replacement of Ca by either Mg at constant Fe or Fe at constant Mg. Zonation in all crystals is weak: <0.5 atomic % Ca (maximum 1.3%) and <1.5% Fe and Mg (maximum 3%).

(v) compositions are comparable with the pyroxene data recorded from other ultrabasic/ultramafic layered intrusions.

2.9.3.2 Minor element variation: The clinopyroxene lattice is capable of incorporating often substantial amounts of minor elements whose distribution and relative abundance are controlled by ionic radius, charge balance and crystal field stabilization energy CFSE (Burns, 1970). Cr, Ti, Al, Mn, Na, K and Ni-contents have been determined for the L.U.C. pyroxenes. Variations result from changing bulk chemistry of the liquid with decreasing temperature, chemistry of coexisting phases, and crystallization order (pressure and rate of cooling are assumed constant during crystal growth).

Cr_2O_3-contents (fig 2.44) are significant and decrease linearly with fractionation from 1.44 at Mg/Mg+Fe 0.89 to 0.1% at Mg-value 0.80. The complete range is shown by both the E.L.S. and C.S.. Comparative values are reported in clinopyroxenes (of similar Mg-value) from the Bushveld (Atkins, 1969; Buchanan, 1979), Salt Lick Creek (Wilkinson *et al.*, 1975) and Jimberlana (Campbell and Borley, 1974) intrusions. Poikilitic pyroxene is weakly zoned (<0.1 wt%), whilst cumulus grains show a strong core to rim decrease (parallel to the fractionation trend) of up to 0.7% in the ol-cpx-plag cumulates, and 0.2% in the plag-cpx-ol cumulates. Within-sample variation is generally outside the limits of analytical error (see Table A.2); reaching a maximum of 0.2 wt% in the peridotites and 0.5 wt% in the allivalites.

Titanium values (fig 2.45) are highly variable, ranging from 0.2-2.05 wt% TiO_2 (in both E.L.S. and C.S.) and increasing linearly with differentiation. Postcumulus grains in allivalite and peridotite show similar variations, and in general the large poikilitic grains are poorer in TiO_2 (<0.7%) than the small isolated intercumulus patches; cumulus crystals contain <1 wt% TiO_2. Such patterns are directly related to the geochemistry of Ti^{4+}, since with its zero CFSE and +4 valency, it cannot compete with Cr^{3+} for charge-deficient octahedral sites, and can only become an important constituent when the liquid becomes sufficiently depleted in Cr^{3+}. Thus early-formed pyroxenes (e.g. poikilitic in olivine cumulates) have high Cr-contents, and middle to late stage pyroxenes (e.g. intercumulus in the allivalites) have high Ti^{4+}.

Zoning of individual grains parallels the overall differentiation trend, and ranges from negligible (<0.05 wt%) when poikilitic, to marked (maximum 1.0 wt% TiO_2 core to rim increase) when cumulus.

Variation between grains is surprisingly large for rocks which are texturally meso- and heteradcumulates (0.2-0.3 wt% TiO_2 in the olivine cumulates and 0.2-0.4% in the plag-ol cumulates) and indicates partial or complete entrapment of liquid by the olivines, and poor diffusional contact (at least for Ti^{4+} ions) between liquid in different pores crystallizing pyroxene.

Al_2O_3-contents of the L.U.C. Cr-diopsides are also relatively high (2.2-4.7 wt%, fig 2.46), and C.S. grains are distinctly richer (3.0-4.7%) than the majority of those in the E.L.S. (2.2-3.5%). As expected, cumulus pyroxene is poor in Al_2O_3 relative to intercumulus, the former having crystallized from liquid depleted in Al (i.e. low activity of Al_2O_3) by the fractionation of cumulus plagioclase. Variation with fractionation is ill-defined, but contents generally show an overall decrease. The trend is more marked in zoned cumulus crystals, which show a sharp core to rim decrease of up to 0.8 wt% (poikilitic pyroxene is unzoned).

Entry of Al^{3+} into clinopyroxene, its relative substitution into tetrahedral or octahedral sites, and its variation with differentiation, are dependent on a number of factors including activities of SiO_2 and Al_2O_3 in the liquid, pressure and temperature changes, and nature of the ion(s) providing charge balance. In the Rhum pyroxene the majority of Al^{3+} is present in the tetrahedral site with Si^{4+} average Al^{IV}:Al^{VI} ratio is 4:1), and since Al^{IV}-contents remained constant during fractionation (activity of SiO_2 therefore constant) the falling Al_2O_3-contents (fig 2.46) must involve decreasing Al^{VI} (octahedral site). Declining Al^{VI}-contents are most directly attributable to decreasing Al-contents of the coexisting liquid (the result of intercumulus plagioclase crystallization). Total pressure is assumed constant during crystallization of the E.L.S. and C.S., and

although it influenced the amount of Al^{VI} entering the pyroxene (increased pressure favours increased Al^{VI} substitution, Verhoogen, 1962; Aoki, 1964), it was not responsible for its variation with differentiation. Textural evidence and the reversed zoning of plagioclase (sections 2.7 and 2.9.2 respectively) suggest that the difference in Al_2O_3-contents between the E.L.S. and C.S. (higher in C.S., fig 2.46) is the result of the latter having crystallized from liquid supercooled with respect to plagioclase (plagioclase supersaturation textures are more widespread in the C.S. cumulates). Such a liquid would have had an unusually high activity of Al_2O_3 and therefore crystallized high-Al^{3+} pyroxenes.

Manganese (fig 2.47) increases slightly with fractionation, 0.09-0.17 wt% MnO in both E.L.S. and C.S., and zoning (parallel to the fractionation trend) is negligible (maximum 0.02 wt% core to rim increase).

Trace amounts of Ni were detected (0.02-0.07 wt% NiO) and minor amounts of Na_2O are present throughout the fractionation range (0.25-0.58 wt%). K_2O was below the detection limit of the microprobe.

2.9.4 Cr-spinel

The compositional diversity of naturally-occurring spinel and its potential as a petrogenetic indicator have long been recognized, and received considerable attention (Thayer, 1970; Irvine, 1965, 1967). However, the physical and chemical factors affecting its crystallization and chemical diversity in stratiform intrusions are still poorly understood, and the subject of considerable debate (Irvine, 1967; Ulmer, 1969; Cameron and Desborough, 1969; Henderson and Suddaby, 1971; Henderson, 1975; Cameron, 1975, 1977; Hamlyn and Keays, 1979). This study describes the compositional variation of the cumulus Cr-spinels, with emphasis on those from the E.L.S., and in the chromitite

layers in particular. The work continues that of Henderson and Suddaby (1971) and Henderson (1975) by considering, in addition, those spinel layers which occur between peridotite and allivalite, and those totally within peridotite.

A wide range of Cr-spinel solid solutions occurs as a result of the extensive substitution of Cr, Al, Fe^{3+} and Ti in the octahedral (B) sites, and Mg and Fe^{2+} in the tetrahedral (A) sites. All Cr is assumed trivalent, and Fe is partitioned between A and B sites in the proportions required for stochiometry (32 oxygens in the structural formula). Representative analyses are listed in Appendix A, and the classification scheme used, illustrated in fig 2.48. The term "Cr-spinel" is non-specific and refers to compositions which contain appreciable amounts of chromium (>15 wt% Cr_2O_3).

2.9.4.1 Primary compositional variation: As discussed in section 2.9.4.2, the Cr-spinels show considerable evidence of postcumulus and/or subsolidus chemical variation, which was superimposed on the primary compositions determined during the initial crystallization (cumulus) stage.

Those spinel grains enclosed by olivine (i.e. inclusion-type) are taken to represent liquidus or near-liquidus compositions, since early entrapment prevented reaction between the spinel and any liquid. However, Irvine (1967) suggested there is evidence, from layered intrusions, of subsolidus re-equilibration between olivine and Cr-spinel, and that the amount of compositional change increases for each mineral as its modal percentage decreases. Roeder *et al.* (1979) ran high-temperature experiments in order to test the degree of redistribution of Fe and Mg between spinel and olivine at subsolidus temperatures, and confirmed that in olivine-rich layers the Cr-spinel is considerably enriched in Fe^{2+} on cooling, whereas the olivine changes only slightly.

Subsolidus re-equilibration, however, does not affect the trivalent-cation contents since Cr, Al and Fe do not readily enter the olivine lattice.

Thus in figs 2.49 and 2.50, the calculated trivalent-cation proportions are assumed close to, if not actual, primary values, and in fig 2.50 primary reaction trends lie parallel to, but an unknown distance from the Mg-rich side of the observed data. Allowing for a small change in Mg/Mg+Fe ratios, the inclusion spinels still classify as chromite and Al-chromite (fig 2.48).

Variation in the trivalent cations (fig 2.49) is limited. The bulk of the data plot in a tight cluster, and the outlying spread may be due to chosen grains not being entirely enclosed by olivine in 3-D, or alternatively becoming enclosed during postcumulus enlargement of olivine by which time they may have reacted with intercumulus liquid. The main variation is in the Fe^{3+}-cation fraction (0.2-0.5) at a more or less constant Cr/Al ratio of 0.7 (a direct result of increasing Fe^{3+}-content of the liquid with fractionation). No zoning of grains has been detected.

Limited chemical variation is further illustrated in fig 2.50, in which the Mg/Mg+Fe ratio is used as a fractionation index (since it shows a positive correlation with that of olivine, see fig 4.19). Ratios range 0.6-0.1. Cr/Cr+Al ratios have a restricted range (0.82-0.48, majority 0.58-0.74) and show an overall increase with fractionation. Comparison with inclusion spinels from other layered intrusions is limited due to paucity of data. Two published analyses from the Elephant's Head Dyke (Eales and Snowdon, 1979) fall within the Rhum field. The Fe^{3+}-cation ratios (fig 2.50) range from 0.1 to 0.5 with decreasing Mg/Mg+Fe. Inclusion spinels from the Elephant's Head intrusion have similar values, but those from the Panton Sill are considerably poorer in Fe^{3+}.

The progressive changes in spinel composition described above, reflect changes in the chemistry of the fractionating magma and may therefore be regarded as a first-order variation.

2.9.4.2 Postcumulus compositional variation: Cameron and Desborough (1969), Ulmer (1969) and Cameron (1975) believed that the precipitation and compositional variation of Bushveld chrome-spinel might be controlled by variations in the magma oxygen-fugacity, but considerable variation of neighbouring spinel grains (see below) is not consistent with this hypothesis. Irvine (1965, 1967) concluded that similar variations could instead result from re-equilibration of Cr-spinel with adjoining silicates during either the immediately post-cumulus stage, subsolidus stage, or both.

Cr-spinels are found within six local environments (excluding those within olivine, described in section 2.9.4.1). (i) within embayments in olivine crystals and surrounded by plagioclase, (ii) within poikilitic feldspar and not immediately adjacent to olivine (in the peridotites), (iii) within poikilitic clinopyroxene (in peridotite), (iv) enclosed by cumulus plagioclase (in the allivalites), (v) between cumulus plagioclase laths, and (vi) enclosed by poikilitic plagioclase in the chromitite layers (discussed in section 2.9.4.3).

Their compositional variations are plotted in fig 2.51 as a function of rock type, and local environment. Only a representative range of data is shown, but several important generalizations are worth noting:

Firstly in the olivine cumulates -

(a) there are relatively slight differences in composition between spinels of a similar environment, in different rocks.

(b) spinel composition in any one rock is a function of the silicate phase enclosing it.

(c) spinels within olivine are the most Cr-rich, and Mg- and Al-poor.

(d) the trends in spinel composition as a function of enclosing phase are not the same for all samples, e.g. there is no consistent order of preference within plagioclase and clinopyroxene for the most Cr_2O_3-rich spinel (compare E51 and M87), and the most aluminous (and magnesian) grains are not always enclosed by the same phase (clinopyroxene in E50 and E51, and plagioclase in M88 and E48). In addition the most Fe- and Ti-rich grains within any one sample can be found in olivine, pyroxene or feldspar (variation in Ti may be due to inclusion of fine-scale ilmenite exsolution, within the microprobe spot).

In the plagioclase cumulates -

(e) spinels between cumulus feldspar laths are consistently higher in TiO_2, FeO, Fe_2O_3 and Cr_2O_3, and lower in MgO and Al_2O_3 than those within the laths.

Compositional variations of the Mg, Cr, Al and Fe^{3+}-cation fractions of spinels from the various environments are shown in figs 2.52 and 2.53. All spinels lie on the same trend, one of increasing Cr/Cr+Al and increasing Fe^{3+}/R^{3+} with decreasing Mg/Mg+Fe, and the complete range of values can be found in a single sample. Spinels range from chromite to spinel (sensu stricto); see fig 2.48. Irrespective of the cumulate assemblage and local environment, the Cr-spinels show the same range in Mg/Mg+Fe ratios, 0.7-0.1. Cr/Cr+Al values are highest for inclusion-types (0.82-0.48), grains within clinopyroxene (0.7-0.5), and grains between cumulus plagioclase (0.65-0.55), and lowest for those within the cumulus feldspar (0.55-0.2). Grains surrounded by poikilitic feldspar show the maximum range, 0.73-0.16. Fe^{3+}-cation fractions range 0.10 to 0.52 in most examples, and up to 0.85 for grains between cumulus, and within intercumulus, plagioclase.

Within any one olivine cumulate sample the most magnesian and aluminous spinels are found within feldspar, and the most chromian

within olivine. In allivalite the most aluminous grains are those trapped within cumulus plagioclase. Those between laths are the most ferric iron-rich.

In a third projection from the Johnston spinel prism (fig 2.54), analyses are plotted in terms of Cr, Al and Fe^{3+}-proportions. Two trends are immediately obvious, and diverge from a common point of origin (lying in the field of liquidus (i.e. inclusion-type) spinels, section 2.9.4.1). One involves an enrichment in Al at the expense of Fe and Cr (the Al-trend), the other enrichment in Fe^{3+} and depletion in Al and Cr (the Fe-trend). They are parallel to the trends defined by Henderson (1975) for disseminated and seam spinels from Units 2, 7, 8, 11 and 12 (see later). Explanation of the trends will be discussed at the beginning of section 2.9.4.3, but at this point it is relevant to point out that the variation ranges far beyond that found within inclusion-spinels. It was concluded in section 2.9.4.1 that the included grains are basically unchanged since the time of inclusion (with respect to trivalent cations), therefore the two trends favour Irvine (1967) and Henderson's (1975) hypotheses of modification of primary compositions by reaction with intercumulus liquid and/or adjacent silicate phases.

2.9.4.3 Variation within chromitite layers: A study was undertaken of the compositional differences across several chromitite horizons, with the aims of (a) extending the work of Henderson and Suddaby (1971) and Henderson (1975) by including additional types of seams, and (b) determining the stratigraphic variation, if any, between specific horizons (i.e. identifying, if possible, a unique chemical "fingerprint" for each seam-type). Six well-developed spinel cumulates were chosen for this purpose. Two are from within olivine cumulates (M87 in Unit 10, and J8 from Unit 12), two occur between peridotite and overlying

allivalite (M77 in Unit 8, and M46 in Unit 11), and two are from the base of cyclic units, i.e. between allivalite and overlying peridotite (KRG at the Unit 11-12 contact, and E1, E19, E30, M68/A from the Unit 7-8 contact). Four samples were taken along a 1 km strike-length of the Unit 7-8 chromitite, to look for possible lateral compositional variation, corresponding to the lateral internal structure variation.

The results of microprobe analyses across these nine seams are shown in fig 2.55. Each analysed grain is unzoned. The following main points can be made:

(i) the degree of compositional variation across spinel layers is a function of their field occurrence. Variation is most marked for those chromitites between allivalite and peridotite (KRG, M68/A, E30, M46), and only slight for seams within peridotite (E1, M87, J8).

(ii) chromitite layers from different environments can show similar compositions (e.g. compare M68/A and J8), and layers from similar environments can show quite different ranges in spinel chemistry (compare M87 and J8).

(iii) the most aluminous spinels are not always spatially associated with plagioclase (i.e. Al-rich) cumulates (e.g. J8),

(iv) the most chromian grains are not necessarily those enclosed by olivine (e.g. E1, E19).

(v) spinel enclosed by feldspar can have a constant composition throughout a sample, irrespective of whether the feldspar is cumulus or intercumulus (e.g. E1).

(vi) individual spinel cumulates cannot be uniquely chemically identified.

In order to illustrate the remarkable range of compositions, and the patterns attributable to the role of the enclosing silicate, and type of liquid (trapped, and non-trapped), each diagram in turn will

be briefly described. Several important suggestions are made regarding the origin of the chemical variation.

Unit 11-12 contact : sample KRG: This chromitite has been studied by Henderson and Suddaby and the observed compositions and variations are similar to those reported by them (1971, fig 2). Compositional variation across the seam involves (going from top to bottom) decreasing TiO_2, Fe_2O_3, FeO, Cr_2O_3-contents and concomitant rises in MgO and Al_2O_3. Analyses of spinels enclosed by olivine in the peridotite are gradational towards those of the uppermost chromitite grains, but there is a marked discontinuity between the basal crystals and those spinels between cumulus feldspars of the underlying allivalite. There are, however, several additional points worth noting, (a) it is significant that all spinels enclosed within plagioclase have similar compositions, e.g. the disseminated spinels of the peridotite resemble the grains in the centre of the seam, (b) those grains in the immediately underlying 1 mm of anorthosite show decreasing titanium, iron and chromium, and increasing magnesium and aluminium towards the seam. This is regarded as indicating possible localized re-equilibration between the two sets of spinels.

Assuming that the inclusion spinels closely approximate to the composition of liquidus Cr-spinel (section 2.9.4.1), the reaction of non-trapped crystals with cumulus phases and intercumulus liquid, appears to have proceeded in the direction of Al- (and Mg-) enrichment. Henderson and Suddaby (1971) and Henderson (1975) suggested the reaction took the form of: ol or plag + Cr-spinel + liq → aluminous spinel + liq (petrographic evidence being the aluminous nature of spinels within olivine embayments, relative to those completely enclosed by olivine). As predicted, reaction with cumulus plagioclase (at base of seam) produced more markedly Al-rich spinels, hence the increase in

Al_2O_3-content towards allivalite. The extent of the reaction was most probably controlled by such factors as diffusion rates of the various elements, length of time intercumulus liquid was present, and proximity to the cumulus phases.

The Fe^{3+}- and Ti-rich grains between cumulus plagioclase laths have reacted, not in the presence of freely diffusing intercumulus liquid, but with trapped residual liquid. A similar conclusion was reached by Henderson and Suddaby (1971).

It remains to be seen if this model fits the rest of the examined chromitite layers.

Unit 7-8 contact : sample M68/A: Identical relationships exist as for KRG. The chromitite shows Al-and Mg-enrichment towards the plagioclase cumulate, and the most Fe- and Cr-rich of the seam spinels are those at the upper contact, which have been poikilitically overgrown by olivine and therefore shielded from subsequent reaction. Disseminated spinels in the olivine cumulate (enclosed by poikilitic feldspar) are similar to those in the centre of the seam, and to those enclosed by cumulus plagioclase.

Unit 7-8 contact : sample E1: This chromitite illustrates well the point just made, that spinels within plagioclase have a constant composition, be they in allivalite,seam,or peridotite. It is tentatively suggested as being the result of subsolidus re-equilibration during slow cooling.

However, trends across the seam are very different from those described in M68/A. Spinels in the upper half of the seam have a constant chemistry, whilst grains in the lower, disturbed part, show a marked chemical variation involving increasing FeO, Fe_2O_3, Cr_2O_3 and TiO_2, and decreasing MgO and Al_2O_3 towards the allivalite (compositionally they resemble grains between cumulus feldspar). This is the

reverse of the normal pattern established in KRG and M68/A, and thought to be directly linked to the disturbed nature of the seam. It is suggested that the lowermost spinels have reacted with trapped liquid whose release from the underlying allivalite may have been triggered by the localized disturbance of the seam. Alternatively, the escaping intercumulus liquid may have disrupted the spinels.

Unit 7-8 contact : sample E30: Although in this case the thin chromitite is developed a few mm above the peridotite-allivalite contact, the within-seam variation is the same as for KRG and M68/A. The trend of increasing Fe_2O_3, FeO, Cr_2O_3 and TiO_2 from the seam, through peridotite and anorthosite to the allivalite, matches that developed in the lower portion of E1 (and KRG, albeit on a smaller scale), and the U-shaped trend-lines illustrate the opposing nature of the Al-enrichment and Fe-enrichment reactions.

Unit 7-8 contact : sample E19: Although this contact is still more complex (having a second thin spinel layer developed in the olivine cumulate) the overall chemical variations and trends are as described above. Spinels within the upper layer are richer in Fe_2O_3, FeO, TiO_2 and Cr_2O_3 relative to those in the lower.

Peridotite-allivalite contact within Unit 8 : sample M77: This is not a true chromitite since the spinel layer contains cumulus olivine grains, but nevertheless is still of considerable interest. Inclusion spinel is slightly richer in iron and titanium and poorer in chromium, aluminium and magnesium in the allivalite than in the peridotite. As in the upper half of E1 there is no compositional variation across the seam, regardless of the type of enclosing silicate, but in contrast to most other samples spinels in cumulus plagioclase are chemically different from those in intercumulus plagioclase, being richer in FeO, Fe_2O_3 and TiO_2, and poorer in Cr_2O_3, Al_2O_3 and MgO.

Peridotite-allivalite contact within Unit 11 : sample M46: Variation patterns are the mirror image of those in KRG, where the lithological relationship is reversed.

Chromitite within peridotite of Unit 10 : sample M87: Although there is no great variation across the seam, Al_2O_3, FeO and Fe_2O_3 show a slight upward increase; Cr_2O_3 and TiO_2 remain approximately constant; and MgO decreases slightly. Spinels in the peridotite above and below are continuous with the seam trend.

Chromitite within peridotite of Unit 12 : sample J8: Within-seam variation is more obvious, and the overall pattern resembles that developed in E30. TiO_2, FeO and Fe_2O_3 remain unchanged across the layer, whilst Al_2O_3 decreases. Cr_2O_3 increases, and MgO increases then decreases. Inclusion spinels above the chromitite are richer in TiO_2, FeO, Fe_2O_3 and Cr_2O_3 than those below. Tie-lines between them are parallel to the seam trend, but the latter spinels are richer in MgO and Al_2O_3 and correspondingly poorer in FeO, Cr_2O_3 etc. The above data provides no evidence for reaction of spinel with trapped liquid.

Variations within the various chromitites in terms of trivalent-cation fractions are illustrated in fig 2.56, with the Unit 7-8 and 11-12 seam trends of Henderson (1975) shown for comparison. M77, the only sample to contain cumulus olivine, is also the only one to show significant variation in Fe^{3+}, 0.22-0.37. The maximum total range shown by the remaining seams is 0.7-0.27, and the maximum for an individual sample is only 0.16 (KRG). Cr/Al ratios show a considerable range from 0.30 to 0.87, and seam trends for the Unit 7-8 contact extend beyond that of Henderson (1975).

A comparison of fig 2.56 with fig 2.54 indicates that, as expected, the seam trends are parallel to and coincident with, the Al-trend shown by disseminated spinel within plagioclase. M77 parallels the

Fe-trend. In figs 2.52 and 2.53 the seam data are coincident with the Mg-rich end (Mg/Mg+Fe 0.4-0.8) of the disseminated spinel trends. It is particularly noticeable that the total range of Rhum spinel compositions extends far beyond those of other stratiform intrusions.

2.9.4.4 Variation between chromitite layers: Samples M68/A, E1, E19 and E30 chosen from various positions along the Unit 7-8 contact (see Map 2) show an east to west increase in iron, titanium and chromium, and a concomitant decrease in magnesium and aluminium. Unfortunately the overlying peridotites lack inclusion-spinels and therefore it is not possible to determine how much of this chemical range is attributable to variation in initial (liquidus) spinel compositions (i.e. changing liquid composition) and how much is a function of differing local environments and extents of postcumulus reactions.

There appear to be no consistent chemical differences between seam spinels from the three main occurrences (within peridotite, at peridotite-allivalite and allivalite-peridotite contacts). Identical spinels are found in very different circumstances (e.g. average J8 and M68/A spinels), and conversely similar environments yield very different spinels (e.g. compare KRG and M68/A). The most aluminous spinels are not necessarily adjacent to Al-rich cumulates, e.g. J8, but the most Fe^{3+}-rich grains occur either immediately above or below an allivalite (e.g. M77, M46 and E1 respectively).

Overall stratigraphic compositional variations are also not apparent, and this is hardly surprising considering the marked compositional modification by postcumulus reactions, and the postulated replenishment theory for the origin of the cyclic units (section 2.8.1).

Variations in trivalent-cation fractions between individual chromitites, are illustrated in figs 2.56 and 2.57. Trends are roughly parallel to one another and to the Cr-Al join, and the extent of Cr/Al

variation ranges from a minimum in M77, to a maximum in KRG. In keeping with the observations in 2.9.4.3 there is no consistent correlation between Cr/Al range and the type, or stratigraphic position, of the chromitite.

Seam trends, together with the data for adjacent disseminated grains are summarized in fig 2.57. Samples J8 and M77 are atypical in that all spinels lie on the same trend (Al- and Fe- respectively). Inclusion spinels which plot a considerable distance along the Al-trend may not be completely enclosed by olivine in 3-D.

In the remaining seven samples, both Al- and Fe-trends are defined by the data. Noticeably, spinels lying within cumulus or poikilitic feldspar consistently plot close to the seam trend (as predicted from section 2.9.4.3). E1, E19 and E30 (all from the Unit 7-8 contact) are very similar, but significantly different from M68/A (also Unit 7-8, but which more closely resembles J8 from Unit 12).

Despite these significant variations, no overall trend is apparent when data are considered in their entirety. By contrast, in successive massive Cr-spinel layers from the Great Dyke there is an increase in total Fe, and decrease in Cr stratigraphically upwards (Worst, 1964). Similarly, in the eastern and western sectors of the Bushveld complex, Cr-contents and Mg/Mg+Fe ratios decrease with stratigraphic height in the layered series (Cameron and Desborough, 1969), and the only systematic stratigraphic between-chromitite change in spinel composition from the Stillwater complex is a steady upward decrease, followed by an increase, in total Fe (Jackson, 1963).

The compositional changes observed within and between Rhum chromitite layers may be considered in terms of a complex interplay of the following parameters: (a) cessation of fractional crystallization (cumulus stage) and initiation of near-equilibrium crystallization in

the crystal pile (postcumulus stage), with postcumulus overgrowth of Cr-spinel occurring in or near the magma-crystal pile interface. Any zonal structure would have been lost in the homogenization process during the slow cooling (Hamlyn and Keays, 1979), (b) effects of postcumulus reaction with intercumulus liquid and nucleating interstitial silicates on settled Cr-spinel, (c) subsolidus re-equilibration between spinel and enclosing silicate.

All three would be superimposed on any initial (primary) chemical variation. Henderson and Suddaby (1971) and Henderson (1975) have presented convincing evidence for the dominant role of (b), and data collected in this study supports their hypothesis. Within-olivine spinel compositions are similar for all chromitites (fig 2.57) and in the light of this, the chemical variations within chromitites can be visualized as the end-product of two competing processes - reaction with trapped or non-trapped liquid - and therefore directly related to variation in the processes of deposition, compaction and diffusion within crystal mushes. The fact that a marked Fe-trend is common within the allivalites, but absent in olivine cumulates, is a reflection of the more efficient trapping of liquid in the former (?a result of faster accumulation rates and rates of plagioclase postcumulus overgrowth), together with the predictably more evolved (Fe and Ti-rich) nature of the magma at the time of allivalite formation.

The role of subsolidus re-equilibration is apparently a function of the local environment, and possibly chemistry, of the spinel. Certainly, the Fe- and Ti-rich spinels between cumulus feldspar laths have not undergone significant Al-enrichment, as would be expected in such an Al-rich environment if this process had occurred. This may, however, be a function of the spinels' high Fe-content (Al being unable to substitute for Fe^{3+}). If re-equilibration has taken place,

spinel within Al-rich feldspar might be expected to have consistently higher Al-contents than grains enclosed by relatively Al-poor pyroxene. This is the case, as shown in fig 2.54, and as already mentioned in section 2.9.4.1, spinel within olivine has been enriched in Fe^{2+}, on cooling. In addition re-equilibration at subsolidus temperatures accounts for the lack of zoning, and the constancy of compositions within plagioclase (either cumulus or intercumulus).

Since similar postcumulus processes might be expected to operate in all layered intrusions, the substantial differences in composition between Rhum spinels and those from other stratiform intrusions (figs 2.52 and 2.53) may reflect differences in parental liquid composition (and hence liquidus spinel and intercumulus liquid compositions) together with differences in intensive variables such as pressure, temperature and oxygen fugacity (Hill and Roeder, 1974).

2.9.5 Ilmenite

The majority of ilmenite in the ultrabasic cumulates is "secondary" or "exsolved", the result of moderate- to high-temperature (>600°C) oxidation of Ti-bearing Cr-spinels (section 2.7.3.7). Ilmenite which crystallized from the liquid as discrete grains ("primary" type) is rare, and restricted to trace amounts (<0.5%) in the plagioclase orthocumulates of Units 6, 7, 8 and 10 (in the former two it is the sole opaque oxide present), and the occasional olivine cumulate (in both E.L.S. and C.S.).

Ilmenite analyses are listed in Appendix A. Total iron-contents (as FeO) have been recalculated into ferrous and ferric, and the analyses into mol % endmember compositions according to Carmichael (1967). CaO and SiO_2-contents are regarded as silicate contamination. Compositions range from $Il_{99.9}Hm_{0.1}$ to $Il_{93.4}Hm_{6.6}$ for both primary and exsolved grains, and are comparable to those from other basic-ultrabasic intrusives (Haggerty, 1976).

Cr_2O_3, Al_2O_3, NiO and MnO minor element contents are low (0.24-1.08, 0.03-0.09, 0.05-0.18 and 0.31-0.89 wt% respectively), and show identical ranges for both primary and exsolved ilmenite, whilst MgO-contents (fig 2.58) are extremely variable and generally high (range 0.49-10.20 wt%, majority >4%). There are no consistent differences in MgO-levels between similar textural types from the allivalites and peridotites, or between textural types from similar rock types.

The two main points to emerge from this data are (i) despite the very low proportion of TiO_2 in the host Cr-spinels (generally <3 wt%), the oxidation product is a magnesian ilmenite, and (ii) despite the widely varying chemistries of the host spinels (section 2.9.4) the exsolved ilmenite shows a very restricted compositional range.

The high-temperature oxidation assemblages of chromian spinels have received very little attention in the literature. Haggerty (1976, p. Hg 74) reports that the spinels only decompose under extreme and intensely-oxidizing conditions, and that for the most part decomposition is restricted to the oxidation of Fe^{2+} and expressed by the development of finely-textured haematite along {111} planes. In the Rhum spinels (and those from the Elephant's Head Dyke, Eales and Snowdon, 1979) it is the high preferential partitioning of Ti into a rhombohedral phase, which dominates the oxidation behaviour of Ti-bearing Cr-spinels, even when the TiO_2-content is <1%. Diffraction data for Rhum (Putnis and Price, 1979) have not revealed haematite, therefore the f_{O_2} evidently did not reach the values required for the formation of haematite.

The absence of oxidation-exsolved ilmenite in those Cr-spinels enclosed by cumulus olivine (despite comparable TiO_2-contents with other spinels) may be due to buffering of the f_{O_2} by the oxidation of the olivine, which shows fine magnetite-pyroxene intergrowths (Putnis, 1979).

2.9.6 Sulphides

The majority of magmatic sulphide deposits lie within the system Fe-Ni-Cu-S. Of the four ternary systems forming the Fe-Ni-Cu-S quaternary, Cu-Fe-S and Fe-Ni-S are the most important with regard to the Rhum sulphides.

The general phase relationships within the much studied Cu-Fe-S system have emerged from the work of Yung and Kullerud (1966), Kullerud *et al.* (1969), Cabri (1973) and Barton (1973). Equilibria at high temperatures (fig 2.59) are dominated by three extensive solid solutions (i) chalcocite-digenite-bornite (cc-dg); (ii) intermediate solid solution (iss); (iii) pyrrhotite (po). As temperature decreases below 600°C, the simple, well-understood phase equilibria gradually give way to less well understood and, in some areas, quite conjectural relationships as additional phases become stable. By far the most important change is the appearance of chalcopyrite ($CuFeS_2$) as a stable phase below 557°C. It forms in the iss-pyrite field and remains isolated from all other Cu-Fe sulphides until the temperature is further decreased. Other important phases to appear are cubanite ($CuFe_2S_3$) at 590°C, and covellite (CuS) at 507°C. Below 500°C the solubility of Cu in pyrrhotite is decreased markedly with declining temperature, and chalcopyrite and pyrrhotite become stable together. Below 330°C (fig 2.60) a series of important changes takes place, including formation of a monoclinic pyrrhotite phase, and the incompatibility of cubanite and pyrrhotite, but a great deal remains to be learned of the phase equilibria at these low temperatures. In summary, minerals that crystallized from the sulphide liquid did not persist during the period of cooling. More or less continual adjustment and re-equilibration took place as temperature (?and pressure) conditions changed. Mineral assemblages occurring presently indicate equilibrium to low

temperatures, among at least some of the sulphide minerals, e.g. chalcopyrite has a 1:1 metal to sulphur ratio at 200°C and the occurrence of low-temperature cubanite indicates equilibrium to some temperature below 210°C.

The general phase relationships within the Fe-Ni-S system have been extensively studied by many workers (Kullerud, 1963; Naldrett *et al.*, 1967; Craig and Kullerud, 1969). High-temperature equilibria are dominated by the $(Fe, Ni)_{1-x}S$ solid solution (known as the monosulphide solid solution, mss), which spans the system from a temperature of approximately 1000°C to below 300°C. Many large-scale deposits, and sulphide droplets in basaltic lavas (Skinner and Peck, 1969), crystallized initially as mss which persisted during considerable cooling, but is rarely, if ever, preserved in nature. As temperature decreases from 1000°C, the mss field shows little change in compositional limits and several binary phases appear, including pentlandite $(Fe, Ni)_9S_8$ at 610°C, until eventually below 400°C mss breaks down (Naldrett *et al.*, 1967). During the cooling the iron-rich portion of mss contracts to pyrrhotite, exsolving pentlandite and pyrite (Craig, 1973), and the limited solid solution towards Cu also contracts by exsolution of chalcopyrite and cubanite (Yund and Kullerud, 1966; Cabri, 1973). The 400°C isothermal section in the Fe-Ni-S system is presented in fig 2.61. Experimental studies of the system phase relations, recently summarized and extended to lower temperatures by Misra and Fleet (1973), imply that the assemblages provide only indirect evidence on the conditions existing when the sulphides collected. Chalcopyrite-pentlandite-pyrrhotite assemblages probably result from chemical adjustment to changing physical conditions down to 230°C. Diffusion of metallic ions within sulphide phases is an important process which will alter original high-temperature histories (Ewers and Hudson, 1972).

A small number of representative analyses of each major sulphide phase are presented in Table 2.1. Samples (B78/1 and KRG) are of the chromitite layer between Unit 11 allivalite and Unit 12 peridotite. All analyses were determined by E.D.S., as outlined in Appendix A.

2.10 Cryptic Variation in the Eastern Layered Series

Within the E.L.S., each layer of olivine cumulate and its overlying plag-ol±cpx cumulate are regarded as complementary products of a single cycle of fractionation corresponding to a single episode of magma influx, with each bimodal unit representing the sequential crystallization: (i) olivine (+ minor Cr-spinel); (ii) plagioclase and (iii) clinopyroxene. This "phase layering", if the result of progressive fractionation of the liquid, should be accompanied by continuous changes in mineral compositions with stratigraphic height ("cryptic layering").

In terms of present knowledge of crystallization of basaltic liquids and of subsequent cooling, the composition of a phase at any given horizon may reflect one, or more, of the following factors:

(a) fractional crystallization of early-formed crystals.

(b) reactions between deposited crystals and supernatant liquid at, or near, the interface between the two.

(c) reactions between deposited crystals and trapped intercumulus liquid.

(d) subsolidus equilibration between coexisting phases.

Three patterns of variation may be found, (i) broad trends upward in the sequence, (ii) medium-scale fluctuations superimposed on the major trends, and (iii) fine-scale variations within the scale of a thin-section, e.g. across chromitite layers (section 2.9.4).

Broad variations may reflect the gross chemical evolution of the

magma by fractional crystallization, whilst medium- and fine-scale variations suggest either the course of differentiation was complicated by intermittent fluctuations of conditions within the chamber, or that compositions have been unevenly modified by postcumulus processes (e.g. (b) → (d) above). In view of factors (b)→(d), caution should be used in identifying possible compositional parameters to indicate significant changes in composition of primary cumulus phases.

Brown (1956) found no appreciable cryptic variation within the E.L.S. cyclic units, and suggested that this implied the maintenance of a relatively constant temperature over a long period of time, as a result of repeated influxes of fresh magma in the early stages of fractionation of each unit. Small fluctuations in the plagioclase compositions were regarded as the result of slight variation in temperature of successive magma batches, or attendant pressure changes, and not differences in the composition of introduced magma.

Wadsworth (1961) however, detected slight base to top decreases in Fo-content of olivines in Units B and D from southwestern Rhum (Fo_{88}-Fo_{85}) and a base to top Fo increase (i.e. reversal) within the thin Transition Member (Fo_{82}-Fo_{86}), based on optical data supported by a few chemical analyses of separated phases.

By the use of electron microprobe analysis, Dunham and Wadsworth (1978) established an unexpected slight (but significant) pattern of cryptic variation in all phases within two selected units (B of the W.L.S. and 10 of the E.L.S.). The most Mg- (and Ca-) rich phases occur slightly above the base of lithological units, whilst the most evolved compositions occur some way below the tops. Compositional trends between units are therefore reversed.

The present study, however, has revealed an even more complex stratigraphic compositional pattern for Units 6-12 from the Barkeval

area. Microprobe analyses have been obtained for the principal minerals in the same samples for which modal and grain-size data are given in Appendix C, Table C.1 (location of traverse is given on Map 2). The data, presented in fig 2.62 indicate that all phases show variations in one or more of their major or minor constituents.

2.10.1 Olivine

Olivine is the dominant cumulus phase and therefore has the potential of offering valuable information regarding the behaviour of the parent liquid. The following generalizations can be derived from the data in fig 2.62. Within the units investigated, the overall range in composition is Fo_{89}-Fo_{79}. Zoning is absent, within-sample variations are small (max. 1 mol% Fo, section 2.9.1), and the average Fo-compositions are plotted. Within the olivine cumulates compositions are confined to a narrow range, Fo_{87}-Fo_{84}, the same for each unit, whilst variation is most pronounced in the thinner plagioclase-olivine cumulates. Once cumulus plagioclase has appeared Fe-enrichment extends the range to ca. Fo_{81} (as far as Fo_{78} in Unit 6). The basal olivines in each unit have dissimilar compositions, ranging from Fo_{84} (Unit 11) to $Fo_{88.2}$ (Unit 12), and there is no systematic variation with stratigraphic height. Indeed, within an individual unit (e.g. Unit 8), the basal olivine can vary randomly along strike (approx. 1 km) from Fo_{85} (J24) to Fo_{88} (E1) to $Fo_{86.5}$ (E19) and Fo_{85} (E30). Compositional differences between olivines at the top of the allivalites and in the immediately overlying peridotites are small (often within the analytical error of the microprobe), e.g. Fo_{84-85} (Unit 7-8 contact), $Fo_{85.5-85.6}$ (Unit 9-10 contact) and $Fo_{85.8-87.4}$ (Unit 11-12 contact). Negligible differences are also found between olivine in the topmost peridotite and the immediately overlying allivalite, e.g. Unit 7, 8, 10, 11. The apparently large difference for

Unit 9 is a function of sampling, the base of Unit 9 was not exposed.

It can be seen that within each peridotite the Fo-content increases sharply within the initial few metres, to a maximum at approximately 2-4 m above the base (e.g. in Units 10, 11). Reversals are also found within the lowermost 2-3 m of each allivalite, and they are more marked, e.g. Fo_{81}-Fo_{85} in Unit 8; $Fo_{82.5}$-Fo_{87} in Unit 10 and $Fo_{84.5}$-Fo_{87} in Unit 11. Above each Fo-maximum (in both peridotites and allivalites) the olivines follow a trend of gradual upward decrease in Fo-content, but in detail the patterns vary from unit to unit, and rock-type to rock-type. In the allivalites of Units 7-10, contents decrease steadily with no sharp reversal to more magnesian compositions as reported by Dunham and Wadsworth (1978) for Unit 10. However, in the top 2 m of Unit 11 such a reversal is found ($Fo_{81.5}$-Fo_{86}).

More complex variations are found within the olivine cumulates. Units 9 and 11 show the simplest patterns: a steady upward iron-enrichment following the initial Fo-increase described above; whilst Unit 8 shows two increasing then decreasing Fo "sub-cycles", the second being the smallest and showing the most marked iron-enrichment (to Fo_{81}). Unit 10, the thickest olivine cumulate, is also the most variable with regard to olivine composition. It appears to consist of three or possibly four "increasing-decreasing" Fo sub-cycles with 1, 3 and 4 involving small but possibly significant changes of 3 mol% Fo (outside the 1% range of within-sample variation), and sub-cycle 2 (the thickest) showing the maximum fractionation to Fo_{81}. Variation in Unit 10 is therefore more marked than that reported by Dunham and Wadsworth (1978).

The recognition of these "sub-cycles" is totally dependent on the small fluctuations in Fo-content (2-3 mol%). It remains to be seen (in subsequent sections) whether these minor compositional oscillations

are mirrored by the chosen parameters for other cumulus and intercumulus phases. One major inference to be drawn from fig 2.62 is that the olivines show no overall progressive "uni-directional" composition change with stratigraphic height.

Olivine NiO-contents (fig 2.62) are highly irregular, but show the same oscillation between narrow boundary limits (0.35-0.15 wt%, majority between 0.35 and 0.25 wt%). Interpretation and possible recognition of subsidiary cycles is complicated by the effects of sulphide-fractionation, e.g. it has been shown in fig 2.41 that olivine of a given Fo-content can have a wide range of NiO-values.

Olivines from the base of successive cyclic units have differing NiO-contents (0.20 wt%, Unit 10 to 0.32 wt%, Unit 11), and although there is no significant overall systematic change with stratigraphic height, those from the uppermost units if anything tend to have slightly higher NiO-values.

In the majority of olivine cumulates the NiO trend parallels that of Fo, but is slightly out of sequence. NiO-inflections occur ca. 4-5 m below those of Fo, e.g. in Unit 8. In the allivalites no such relationship is evident. Only in Units 9 and 10 does NiO parallel Fo, and in the expected sense of fractionation. In Unit 10 there is no initial basal NiO-reversal corresponding to that of Fo, and in the remaining allivalites (Units 6, 7, 8, 11) NiO and Fo trends are antipathetic.

There are no major jumps in NiO, of olivines between peridotite and overlying allivalite, e.g. in Units 7, 8, and 11 (exception being Unit 9, due to incomplete sampling), but between the top of one unit and the base of the next there are often very marked NiO-reversals, e.g. an increase at the Unit 8-9 and 10-11 contacts, and a slight decrease between Unit 7 and 8. There appears to be no change across the Unit 9-10 contact.

2.10.2 Plagioclase

Plagioclase shows a less restricted range of compositions than olivine (An_{90}-An_{72}, section 2.9.2), with the intercumulus grains showing the maximum variation. Within-sample variation is generally <2 mol% An, and therefore average compositions are plotted for each sample. Where cumulus plagioclase is present (in the allivalites) there are several trends and relationships with associated olivine.

(i) in Unit 6 the feldspar shows Ca-enrichment (An_{76}-An_{82}) paralleling, but more pronounced than, Mg-enrichment in the olivine

(ii) Unit 7 feldspars show marked An increase (An_{84}-An_{90}), but are accompanied by slight Fe-enrichment in the olivines

(iii) In Unit 8 Na-enrichment (An_{82}-An_{73}) is associated with a marked increase, then decrease, in olivine Fo-content

(iv) In Unit 9 allivalite, it is the feldspar which shows the reversed, then normal, compositional change

(v) feldspar and olivine compositions trend in the same sense in Unit 10, both showing an initial reversal, followed by normal Na- and Fe-enrichment. Olivine shows a more pronounced change than the plagioclase

(vi) Unit 11 resembles Unit 8 in that the "reversed" plagioclase fractionation accompanies a "normal" olivine trend.

There appears to be no systematic correlation between type of feldspar trend and stratigraphic height, but in the sequence Unit 8-11, the overall Ca-content of cumulus plagioclase generally increases. The composition of the basal feldspar is only slightly different in each unit, varying An_{79}-$An_{84.5}$-An_{86} with increasing stratigraphic height.

Intercumulus plagioclase compositions show an equally complex stratigraphic variation. Discounting obvious marginal zoning, the

range of within-sample variations is larger than for the cumulus grains (ca. 6 mol% An, section 2.9.2), and therefore for each sample bars indicating the full range are plotted, together with the average core composition. From fig 2.62 it is evident that there is little recognizable similarity of the intercumulus feldspar to the cumulus olivine trend. Only in Unit 9 peridotite (and possibly Unit 8, if sample M72 is atypical) does the plagioclase show a normal fractionation trend of increasing Ab-content (An_{80}-An_{73}). Elsewhere the tendency is to show an overall increase in An with increasing height. This is best shown in the higher units (10 and 11). In the former the trend is interrupted by a sudden jump to more sodic compositions (ca. 2/3 way up the olivine cumulate), but does not correspond to any significant contemporaneous change in olivine composition.

Between cyclic units the feldspar (unlike olivine) shows large compositional differences. In each case the poikilitic grains at the base of one unit are markedly less calcic than the cumulus grains of the underlying allivalite, e.g. An_{90}-An_{85} across the Unit 7-8 contact, and $An_{86.5}$-An_{79} across the Unit 9-10 contact.

In contrast the differences between feldspar in peridotites and overlying allivalites are negligible (e.g. An_{85}-An_{84} in Unit 7). The large jump from An_{76}-An_{85} in Unit 9 may be a function of incomplete sampling across the poorly-exposed peridotite-allivalite boundary.

There is no systematic variation in the type (reversed or normal), or degree, of cumulus and intercumulus zoning, with stratigraphic height.

2.10.3 Clinopyroxene

The compositional record for clinopyroxene is less complete than for plagioclase and olivine, due to its scarcity in many of the sampled allivalites. Cumulus grains have only been sampled from Unit 8. It is

clearly shown in fig 2.62 that on the whole the pyroxene Mg/Mg+Fe trend parallels that of the cumulus olivine (despite minor discrepancies, e.g. at base of Unit 9), occurs within a similarly restricted range (majority between 0.85 and 0.88), and bears no resemblance to the intercumulus feldspar trend. Cumulus pyroxene in Unit 8 shows a reversed trend, continuous with the overall trend for that unit. As with olivine, there is no significant progressive up-sequence change in composition, and as with plagioclase there is often a jump to more fractionated compositions at many inter-unit contacts, e.g. 7-8, 8-9, 9-10.

Due to its rapid exponential fall in clinopyroxene with differentiation (section 2.9.3.2), Cr_2O_3 should prove a more sensitive parameter for the identification of fractionation trends and subsidiary cycles. The Cr_2O_3 variation identifies numerous sub-cycles which correspond to those tentatively suggested from subtle variations in Fo-content of the olivines (2.10.1), e.g. Unit 8 olivine cumulate contains two sub-cycles of decreasing Cr_2O_3-contents, separated by a sudden "resetting" to high Cr_2O_3-values, and Unit 10 peridotite comprises three reversals (at M84, M86 and E49). Resetting to higher Cr_2O_3-values also occurs at the base of each allivalite, and is most pronounced in Unit 8.

2.10.4 Cr-spinel

As shown in section 2.9.4, the disseminated Cr-spinels show considerable chemical variation as a result of postcumulus reaction processes involving the spinels, adjacent silicates, and the intercumulus liquid. In order to minimize the modification of any primary cryptic variation by such processes, only the grains totally enclosed within olivine have been plotted. Zoning is absent, and variation between grains in any one sample is less than that shown by grains

from different stratigraphic levels. Data are scarce for the allivalites due to the relative paucity of spinel and olivine in these rocks. The Mg/Mg+Fe ratio is not used since its variation within a sample is greater than that between samples.

Cr_2O_3-contents (fig 2.62) almost exactly parallel the clinopyroxene Cr_2O_3-trend, but are slightly out of phase (the spinel inflections occurring on average 5 m down-sequence from those in the pyroxene). There is no consistent up-sequence change between units; spinels in higher units are just as Cr_2O_3-rich as those lower in the succession. It should be noted that spinels from the plag-ol cumulates do not differ significantly from those in the olivine cumulates.

Compositional differences between the various chromitite layers have been discussed in section 2.9.4.3.

2.10.5 Discussion and conclusions

From the data presented in section 2.10 there is clearly slight, but significant, cryptic variation within the E.L.S.. The pattern is broadly linked to the repetition of the macro-units, and is taken to support a multiple-intrusion hypothesis (section 2.8.1), with the upward sequence of phase and cryptic layering in each unit being the result of progressive in situ fractionation of the magma. However, in contrast to the contention of Brown (1956) and Wadsworth (1961) that each macro-unit corresponds to a separate batch of magma, data presented in sections 2.10.1 (NiO-contents of olivine) and 2.10.3 (Cr_2O_3-contents of clinopyroxene) suggest that each unit in fact comprises the crystallization products of more than one influx of magma; 3, 2 and 3 (possibly 4) subcycles have been tentatively identified in Units 8, 9 and 10 respectively. Each sub-cycle consists of a "reversed" and "normal" trend, with the former representing a period of mixing between the residual magma of one injection episode and the

succeeding fresh magma batch (occurring on a limited scale - reversals average 10-15 m thick - due to replenishment at an early stage when the effects would be less dramatic).

However, having interpreted the NiO and Cr_2O_3 cryptic variation patterns as primary stratigraphic features, the possibility must also be considered that part, if not all, of the variation is the result of the modification of primary compositions by postcumulus (secondary) processes. For example, the fresh magma might be expected to have exerted a chemical influence on the intercumulus liquid in the relatively unconsolidated material in the uppermost few metres (ca. 10) of the crystal pile, such that the accumulated phases re-equilibrated with the hotter, more primitive magma. However, textural evidence of such a process having occurred is unlikely to have survived subsolidus re-equilibration during slow cooling.

Although intercumulus liquid most probably played an important role in the modification of mineral compositions, the extent and nature of such changes cannot be fully assessed due to (a) the complex, and as yet not fully understood mechanisms of crystal-liquid segregation and movement through a cumulate pile, (b) the possibly only transitory existence of the intercumulus liquid(s), and (c) the unknown composition of the liquid(s).

Since the presence of similar compositional ranges and patterns in each macro-unit, and the lack of systematic variation with stratigraphic height imply repeated batches of uniform magma, the magma is unlikely to be the product of a deeper-level fractionating magma chamber. The apparent slightly differing olivine compositions (Fo-content) at the base of each unit is seemingly at variance with this conclusion, but may be a function of the secondary modifications outlined above.

The marked increase in An-content of the intercumulus (and majority of the cumulus) plagioclase with stratigraphic height, both between and within units, cannot be satisfactorily interpreted in terms of the replenishment and magma mixing model, since the feldspar cryptic variation pattern bears no relation to those of olivine, clinopyroxene and Cr-spinel. However, Yoder *et al.* (1957) demonstrated that an increase in P_{H_2O} in the An-Ab system will lower the liquidus and solidus, resulting in a more calcic plagioclase crystallizing. Evidence for a progressive increase in P_{H_2O} in successive magma influxes is suggested by the close association between plagioclase reversals and the development of harrisitic texture (high water-contents are necessary for harrisite formation, Donaldson, 1975a); the reversals are most strongly developed in those portions of the sequence which are harrisite-dominated, e.g. in Units 8, 9 and 10.

The nature of the magma responsible for the E.L.S. must also be considered, and arguments centre around the MgO-content (basaltic, <9 wt% versus picritic, >9 wt%) and olivine phenocryst-content. Dunham and Wadsworth (1978) regarded it as unnecessary to invoke picritic compositions, concluding that a basaltic liquid is capable of producing the established fractionation pattern and the mineral compositions concerned. 1 atm. melting experiments (Forster, 1978) confirm that a tholeiitic basalt (Brown, 1956, Table 8, Anal. 1) will crystallize the sequence olivine, plagioclase, clinopyroxene as required by the cumulus sequence in each macro-unit. However, the results do little to resolve the problem of the limited degree of cryptic variation, in that (a) there is a considerable drop in temperature (110°C) from the incoming of olivine to the appearance of clinopyroxene on the liquidus, and (b) the liquid composition may crystallize insufficient olivine to account for the thick olivine cumulate sequences.

To overcome this difficulty of substantial falls in temperature in the magma chamber, Gibb (1976) suggested the parental magma arrived in the Rhum depositional chamber, not as a basaltic liquid, but rather as a highly olivine-phyric (and spinel) picrite or high-MgO basalt, and at a temperature close to the plagioclase liquidus. This hypothesis, however, appears inconsistent with the pattern of cryptic variation now known to exist in the Rhum E.L.S., since it is highly unlikely that crystal settling from an olivine suspension would lead to any systematic variation in olivine composition with stratigraphic height (within each unit). Since the only fall in temperature within the chamber would be from the onset of plagioclase crystallization (ca. 80°C, Forster, 1978), it could be predicted that cryptic variation will be more substantial in the allivalites than the peridotites. Data in fig 2.62 weakly support this. If Gibb's (1976) hypothesis is correct, the observed compositional reversals within the peridotites are not a primary stratigraphic feature, but the result of post-cumulus modification.

An alternative replenishment model is presented, which accounts for (a) the limited degree of cryptic variation, and (b) the rather constant end-point of each macro-unit (replenishment always immediately follows the appearance of clinopyroxene as a cumulus phase).

The parental liquid of the Rhum cumulates is believed to be of picritic (high-MgO, >9 wt%) composition, similar to that proposed for the dyke suite (see Chapter 6). In their fluid dynamical investigation of the replenishment of a basaltic magma chamber by the influx of hot, dense, picritic liquid, Huppert and Sparks (1980b) proposed a general model which has several important geological applications and provides an explanation for the bimodality of large-scale cyclic layering in ultrabasic intrusions. A feature of the model is that

the picrite does not immediately mix with the basalt, but spreads over the chamber floor, forming an independent layer a few tens of metres thick. This picritic layer convects vigorously as it exchanges heat with the overlying basalt across a stable interface.

Huppert and Spark's (1980b) calculations show that the convective velocities of the crystallizing olivines are far greater than their settling velocities, and thus predict that the picrite layer develops into a suspension of olivine crystals in a liquid which evolves towards a basaltic composition as the picrite cools to thermal equilibrium with the upper (basaltic) layer.

The most important implication of the model is that the growth of olivine in a well-mixed convecting layer (i.e. under conditions of equilibrium crystallization) would result in little or no compositional variation in the resulting cumulate layer: a feature already noted in the Rhum peridotites.

The model also suggests the reason for the constant end-point of each Rhum macro-unit. As the two layers of liquid (influx and resident) approach thermal equilibrium, convection in the lower layer dies down, olivine begins to settle, and the residual liquid (likely to be close to the density minimum with 9-11 wt% MgO - Huppert and Sparks, 1980b, fig 4) becomes buoyant, segregates and mixes convectively with the overlying basalt. At this stage plagioclase and then clinopyroxene join olivine as liquidus phases, and plag-ol±cpx cumulates (allivalites) are generated until interrupted by the next fresh influx of picrite magma. Since conditions of vigorous convection and equilibrium crystallization no longer exist, the allivalites show more marked fractionation and cryptic variation than the peridotites. Conditions of magma influx, storage, and removal were such that each influx of picrite attained thermal equilibrium with the resident basalt before the next replenishment.

In conclusion, the cryptic variation data are taken to support a magma replenishment model in which each macro-unit is the consequence of the influx of picritic magma into a basaltic magma chamber. NiO (olivine) and Cr_2O_3 (spinel and clinopyroxene)-reversals are assumed to be primary stratigraphic features, and imply several small magma influxes during the early stages of some replenishment cycles (e.g. Units 8, 10). Superimposed on the An- and Fo-reversals are the effects of the build-up of P_{H_2O} in the magma, and modification by post-cumulus processes. Estimates of MgO- and phenocryst-contents of the parent magma are suggested in Chapter 6 and section 2.9.1 respectively.

2.11 Summary of the Main Points

1. The Rhum layered ultrabasic complex comprises three layered series: the Eastern Layered Series (E.L.S.), Western Layered Series (W.L.S.) and Central Series (C.S.).
2. Only Units 4-12 of the E.L.S. type-section continue westwards into the Barkeval-An Dornabac area.
3. The W.L.S. is re-defined as consisting of the Harris Bay and Ard Mheall Members.
4. The newly-defined C.S. consists of the Outer Breccia, Dornabac, Long Loch and Ruinsival Members. It has discordant intrusive and faulted contacts against the E.L.S. and W.L.S., and at its northern and southern extremities extends across the Inner Ring Fault, thereby post-dating the emplacement of the E.L.S. and W.L.S.
5. The Long Loch Fault appears to have played a significant role in the distribution of the C.S.
6. The structural relationship between the E.L.S. and W.L.S. is unresolved.

7. Olivines range $Fo_{89.4}$-Fo_{78}. In the E.L.S. they have equilibrated with a sulphur-saturated magma; in the C.S. with a sulphur-undersaturated magma.
8. Ni-sulphide concentrations are predicted at depth in Rhum.
9. In the E.L.S., sulphide concentrations (magmatic) are associated with the Cr-spinel cumulates: Cu-Fe and Fe-sulphides are dominant (cp, bn, po, cb), with minor Fe-Ni and Cu-sulphides (pn, cc, cv).
10. Plagioclase ranges An_{90}-An_{53} (including zoning), but clinopyroxene is restricted to diopsidic compositions (Cr- and Al-rich).
11. Cr-spinel cumulates (chromitites) are best developed in the E.L.S. and are found at the base of many cyclic units, at numerous horizons within the peridotites, and at the base of several allivalites.
12. The origin of the macro-rhythmic layering (and chromitites) is explained in terms of multiple intrusion, i.e. the repeated influx of fresh (MgO-rich) magma into the depositional chamber.
13. Fine-scale rhythmic layering in the 2 and 3-phase cumulates is probably the result of gravitational deposition from moving currents. In situ crystallization is more likely to have occurred within the olivine cumulates.
14. Cryptic variation within the E.L.S. (both cumulus and intercumulus phases) is more complex than previously thought, and the pattern supports a magma replenishment model.
15. The recognition of "sub-cycles" within the cyclic units implies several small magma influxes during the early stages of some replenishment cycles.

CHAPTER 3

THE GABBRO MINOR INTRUSIONS

3.1 Introduction

The term "gabbro" as used in this study, refers to coarse-grained igneous rocks composed principally of plagioclase (>An_{50}) and pyroxene (usually cpx, but can be cpx + opx) with or without olivine. In the Rhum context such rocks lack layered structure, i.e. are devoid of modal (rhythmic) layering and feldspar lamination.

The terms "eucrite" and "gabbro" have featured widely in the Rhum literature (section 3.2). In view of the very varied history of the term "eucrite" (Le Bas, 1959), and the confusion and inconsistency associated with its use on Rhum (Harker, 1908a; Brown, 1956; Wadsworth, 1961), all coarse-grained basic minor intrusions on Rhum will hereafter be referred to as gabbros.

3.2 Previous Research

The majority of the gabbroic minor intrusions occur within the Main Ring Fault where they are intimately associated with the layered ultrabasic rocks and demonstrably younger than them. To date, approximately twenty-one gabbro masses have been mapped (Harker, 1908; Brown, 1956; Hughes, 1960b; Wadsworth, 1961; Dunham, 1962), ranging in size from small dyke-like masses a few metres wide by tens of metres long, to large masses measuring up to 400 x 700 m (fig 3.1). In form the intrusions vary from vertical dykes (Fiachinis) and elongate dyke-like bodies (Papadil, Loch Gaimlich, Barkeval) to 0.5-30 m thick sheets concordant with (Glen Duain, Ruinsival), or transgressive to (An Dornabac, Hallival, Barkeval), the Layered Series. In addition irregular bodies of gabbro intensely vein the ultrabasic rocks (Askival, Barkeval). In general, the gabbro intrusions are concentrated east of

the Long Loch Fault (fig 3.1).

Harker (1903) described and mapped the majority of the gabbroic intrusions (including those distinguished as "eucrites") but they received no further attention until the reinterpretations suggested by Bailey (1945) and the petrological research of Wager and Brown (1951) stimulated major reinvestigations of the igneous rocks. This new phase of mapping by such workers as Brown (1956), Wadsworth (1961), and Dunham (1962) reinterpreted some of Harker's "eucrites" and could add only a few new gabbroic intrusions to the Rhum map.

Working in the Hallival-Askival area, Brown (1956) showed most of the strip originally mapped by Harker (1908) as "eucrite" sheets, to be the three lowermost units of the Eastern Layered Series, and the two allivalite sheets to be post-ultrabasic intrusions of fine-grained olivine gabbro in the form of slightly transgressive inclined sheets. Brown (1956) also confirmed Bailey's (1945) interpretation that these two fine-grained gabbro sheets join up with a marginal gabbro believed to be in the form of a ring dyke. Other newly mapped post-ultrabasic gabbros included several steeply inclined sheets on the Hallival-Barkeval col.

Wadsworth (1961) recorded an even smaller volume of gabbroic material in the Western Layered Series. A large "eucrite" body was reinterpreted as the Harris Bay Member, but the boundaries of the An Dornabac, Glen Harris and Papadil intrusions remained largely unchanged from Harker's original map. The only new gabbros to be found by Wadsworth (1961) were the conformable sheet on the north-west face of Ruinsival, the Fiachinis "dyke" and a number of thin conformable sheets in the Glen Duain section of the Harris Bay Member.

When the complex northern margin of the Rhum igneous centre was mapped by Dunham (1962, 1968) field relationships indicated at least

two ages of gabbro within the Main Ring Fault: the "early" gabbros which contributed to the explosion breccias, and the "late" gabbros which cut these breccias together with the ultrabasic tongues near Priomh Lochs. Thus the "early" gabbros represent the earliest phase of basic magmatism on Rhum.

Gabbro intrusions, however, are not confined to the main igneous complex. Plugs varying in size from 20 m to 50 m maximum diameter are abundant in the Torridonian arkoses of NW Rhum (up to 2 km from the Main Ring Fault), as first noted by Harker (1908), and in the arkoses just outside the Main Ring Fault in the Cnapan Breaca - Long Loch area (Dunham, 1962; 1968). The general NW-SE elongation of these plugs is parallel to, and coincident with, the major axis of dilation defined by the Rhum linear dyke swarm (Speight, 1972) and is undoubtedly a reflection of the regional stress pattern. Despite their location in areas of known high dyke density, the plugs are cut by very few dykes and therefore probably post-date the main linear dyke swarm (Forster, 1980).

Because of poor exposure, the tracing of gabbros across the Main Ring Fault is difficult and correlation of the gabbros inside and outside it can only be tentative. Dunham (1968) suggested that the altered gabbro plugs outside the Main Ring Fault may be correlated with the "early" gabbros within. However, petrographic evidence, presented later, makes correlation with the "late" gabbros more probable.

In summary, the gabbroic intrusions of Rhum have received relatively little geological attention. Descriptions of field characteristics and petrography have been given by Harker (1908a), Brown (1956), Wadsworth (1961) and Dunham (1962) but very little attention was paid to the genetic relationship, if any, between the gabbros (and "eucrites") and the ultrabasic rocks. However, Brown (1956) concluded that the Barkeval

"eucrite" showed more marked mineralogical affinities with the ultrabasic rocks than did any other gabbro, and may have formed from interstitial liquid filter-pressed from the layered rocks. Similarly Wadsworth (1961) suggested that the magma from which the gabbros crystallized was closely related to the parent magma of the layered series. Despite the probability of a genetic link between the two groups very few geochemical studies were undertaken, and Brown (1956) presented only two whole rock analyses of gabbros, noting their similarity to his calculated "layered series" parental magma composition. In general the gabbroic and ultrabasic rocks have been regarded as two distinct and separate groups.

Because of this paucity of data a detailed study of the coarse-grained basic igneous rocks was undertaken with a view to a) describing their mineralogical and chemical variation, b) establishing chronological relationships and c) ascertaining the genetic relationship, if any, between the gabbroic and ultrabasic rocks.

3.3 Field Relations

The ultrabasic rocks of the Barkeval-An Dornabac region are invaded by a number of gabbroic intrusions. Some twenty-one have been mapped within the study area, of which fourteen are new (see fig 3.2). All are demonstrably younger than the layered ultrabasic rocks and take the form of plugs, inclined sheets or irregular networks of veins.

The plugs are generally elliptical in outcrop and form low rounded knolls whose dark grey colour contrasts with the pale brown feldspathic and dunitic peridotites. They vary in size from 5 x 2 m to 800 x 200 m. Contacts against the ultrabasic rocks, where seen, are approximately vertical. The majority of the plugs have veined

and enclosed fragments (<10 cm → >1 m) of the surrounding layered ultramafites, thus forming intrusion-breccias. Very few plugs contain ultrabasic fragments throughout their length and breadth. Depending on their size, the plugs can be homogeneous or heterogeneous. The smaller plugs tend to be very homogeneous in that they are even-grained, show no variation in mineral proportions, and rarely contain xenoliths of the country rock. Olivines "weather in" as brown pits, while the coarse ophitic and subophitic dark green pyroxenes "stand out" amongst the dark grey feldspar laths. Those gabbros with a slightly finer grain-size and more granular pyroxenes are smooth-surfaced.

In contrast the larger plugs are very heterogeneous. They are somewhat variable in grain-size and mineral proportions, typically showing irregular streaks and patches of finer-grained gabbro enclosed in coarse-grained varieties. Occasionally they contain pegmatitic patches (average grain size >2 cm). The streakiness is usually attributable to variation in olivine content (varying from a few per-cent to 40 percent over a distance of a few centimetres) and the irregular olivine-rich bands and lenses parallel the intrusion margins.

The two large gabbro masses on the western and southern faces of Barkeval are especially complex. On the western face, an elliptical plug of homogeneous grey gabbro becomes increasingly heterogeneous eastwards with the progressive introduction of streaks and lenses of a brown, finer-grained, more olivine-rich gabbro. In places this brown gabbro veins the grey variety, and vice versa. Other exposures show a very feldspathic variety of gabbro in which the small percent of reddish-weathering olivines gives an orange-brown colour to the rock (not unlike some olivine-poor allivalites in the E.L.S.), and pyroxene is inconspicuous. Along its northern margin the streaky

grey-brown gabbro has veined and brecciated the E.L.S.; angular blocks (up to 2 m across) of allivalite and peridotite are randomly mixed and distributed throughout the resulting intrusion breccia. In a broad zone along its eastern and northern margins this intrusion-breccia is, itself, veined and brecciated by brown-weathering basic material varying from fine-grained dolerite to coarse-grained picrite. The resultant polymict breccia (fig 3.3) consists of blocks of allivalite and feldspathic peridotite (some showing the first-stage brecciation by the heterogeneous gabbro) together with several varieties of gabbro (representing the variation within the plug), randomly mixed and enclosed in a fine-grained, slightly olivine-phyric basaltic matrix. A local variation of this polymict breccia (fig 3.4) shows the fragments to be much smaller (averaging 10 x 5 cm), wedge-shaped, and defining a "bedding", with a faint ?imbricate structure, dipping 30° east. Further west, where the fine-grained basic material has brecciated the gabbro "plug proper", the resultant breccia is monomict, containing closely-packed angular fragments of grey gabbro only (varying from 2 cm → 0.5 m).

Along the eastern margin of the plug the invading basic material is picritic (containing >50% olivine phenocrysts). Contacts are sharp but lack chills. This picritic material often contains small subrounded fragments of feldspathic peridotite and allivalite, and nowhere can it be seen to have invaded the adjacent Ruinsival Member on western Barkeval (see Map 1). Hence the latter may post-date the brecciation of the gabbro.

Following brecciation by this basic material (both olivine-poor and olivine-rich varieties), the whole plug complex was intruded by two cross-cutting generations of dykes (see Chapter 4).

An identical story can be deduced from the field relationships of the large gabbro mass exposed in the cliffs on the lower section of the south face of Barkeval (see Map 1).

Beside the river in Atlantic Corrie are several large outcrops of a very leucocratic homogeneous gabbro. It too is invaded by veins and patches of picritic material and a swarm of dykes, but texturally and mineralogically it is very different from the heterogeneous gabbro exposed in the cliffs above. The pale whitish weathered surface has a distinctive "spotted" appearance due to the presence of green poikilitic pyroxene crystals. Harker (1908b) mapped this leucogabbro as an allivalite. This is not surprising since the gabbro is very difficult to distinguish from some of the poorly-layered allivalites in the Eastern Layered Series. Similar textures have been recorded from the Anorthosite Zone of the Kalka Layered Intrusion (Goode, 1977), in "mottled" anorthosites from the Bushveld Complex (Cameron, 1969) and from anorthositic rocks in Labrador (Phinney, 1967; Emslie, 1970).

None of the gabbros mapped in this study shows chills, suggesting their intrusion took place soon after the consolidation of the layered ultrabasic rocks. All those within the layered ultrabasic complex are fresh compared to the highly chloritized and epidotized gabbro masses ("early" gabbros, Dunham, 1968) bordering the northern margin of the complex.

In contrast to the formation of steep-sided plugs, gabbro is abundantly present in the form of vein stock-works which can cover an area up to 400 x 300 m. Three types of vein can be distinguished in the field.

Type (a): are characterized by a high percentage of mafic minerals, chiefly pyroxene and Fe-Ti oxides. They have a very limited distribution within the L.U.C., occurring as occasional veins in Unit

8 of the Barkeval portion of the Eastern Layered Series near the Triangular Loch, and as small local concentrations of veins in the Ruinsival Member peridotite on western Barkeval. Brown (1956) described similar gabbro, the "Askival Plateau Gabbro", as veins in Unit 10 of the Eastern Layered Series.

Type (b): consist of dark grey, often streaky gabbro similar in grain size, texture and olivine-content to the large plugs described previously, and occur mainly around the summit of Barkeval where they brecciate the layered rocks.

Type (c): are the most widespread and abundant type, consisting of very coarse-grained gabbro with large (1-2 cm) crystals of green pyroxene and white feldspar with subsidiary smaller olivines (2 - 3 mm). These veins, ubiquitous throughout the peridotites, average 4 cm in thickness, and are usually inclined at a low angle to the igneous layering. Similar examples have been defined by Brown (1956) as "contemporaneous ultrabasic veins".

This simple field classification of the gabbro veins corresponds to textural, modal and mineralogical variations revealed by petrographic studies of thin sections (section 3.4).

In conclusion the gabbroic minor intrusions are younger than the layered peridotites and allivalites, are generally dark grey in colour, and lack the continuous internal igneous layering typical of the allivalites. Although the majority fit this field definition, a few gabbros show textures and mineral proportions more characteristic of olivine- and pyroxene-rich allivalites.

3.4 Petrography

3.4.1 Nomenclature

A rational scheme of nomenclature based on the proportions of Ca-rich and Ca-poor pyroxene, and on the presence or absence of olivine will be used in the following descriptions. Viz: (a) gabbro (plag-cpx); (b) olivine gabbro (plag-cpx-ol); (c) hypersthene gabbro (plag-cpx-opx); (d) olivine hypersthene gabbro (plag-cpx-opx-ol). Classification in the field is generally based on the presence or absence of olivine only, as orthopyroxene is present in minor amounts (<5%) and therefore difficult to identify.

However, a classification based solely on modal compositions is not sufficiently discriminating, since there is considerable overlap both between the four gabbro types and between the "non-layered" gabbros and the "layered" ultrabasic rocks (section 3.3). Any subsequent petrographical classification must, therefore, involve textural parameters.

3.4.2 Textures

Textural variations emphasize the interpretational problems which can arise from trying to apply either "cumulate" terminology (Wager *et al.*, 1960) or a standard "non-cumulate" textural nomenclature to a single series of ultrabasic to basic rocks. A non-cumulate classification will be used in this study; based on the texture of the pyroxenes. Four main varieties of pyroxene - discrete, ophitic, intergranular and granular - together with their subvarieties and intergradational relationships, are summarized in fig 3.5.

The definition of "ophitic" follows that of Walker (1957) and refers to subhedral or anhedral pyroxene in optically continuous areas and completely enclosing plagioclase laths whose average length is less than that of the pyroxene. If plagioclase is only partly enclosed the

term "subophitic" is used, and with a further decrease in pyroxene-content the pyroxene occurs in isolated but optically continuous areas and the texture is said to be "neophitic" (Walker, 1957). If the areas are not in optical continuity the pyroxene is "interstitial".

Those gabbros with pyroxene as the sole or dominant mafic phase show a similarly gradational suite of textures. When the pyroxene forms aggregates of large subhedral crystals, free of plagioclase laths, and with a grain size greater than that of the feldspar, such that the rock takes on a "porphyritic" appearance, the texture is described as "discrete". If the crystals marginally partly enclose plagioclase the texture is transitional towards "subophitic". Where the subhedral crystals are of a similar size to the feldspars and arranged in a network that may be random or subradial or subparallel, the gabbro has an "isogranular" texture (after Oppenheim, 1964). When the pyroxene grains are smaller than the feldspars and situated between them, the texture is "intergranular" (Oppenheim, 1964).

Fig 3.5 illustrates the point, already made, that the gabbros are often transitional in texture towards the layered peridotites and allivalites. The large "ophitic" pyroxenes of plagioclase-rich gabbros (e.g. B88) can closely resemble the "poikilitic" pyroxenes of the "mottled" plagioclase cumulates, and similarly, the subophitic, neophitic and intergranular textures can, if present in more olivine-rich gabbros (e.g. B7, E77A) closely resemble the "subpoikilitic" texture of the layered peridotites. The close similarity between "discrete" and "isogranular" gabbros (E67 and M7) and the pyx-plag-ol cumulates of the Dornabac Member (B22 and B1) - figs 3.6 and 3.7 - further illustrates the textural relationship between so-called "cumulate" and "non-cumulate" rocks.

The interpretation of textures as either "cumulate" or "non-cumulate" is therefore open to criticism. If the gabbro plugs and sheets, which in the field display no evidence of igneous layering, are assumed to have crystallized virtually in place and to approximate in composition to the liquid form which they crystallized, then their anomalous "cumulus" textures must be interpreted in terms of nucleation and growth, rather than rates of crystal accumulation and equilibration with the parental magma. However the possibility must also be considered that these gabbros resulted from crystallization of basaltic (or "picritic") magmas containing variable percentages of phenocrysts. On the macro-scale a rock may show igneous layering and therefore be interpreted as a cumulate, but on a micro-scale it may show a "gabbroic" texture. Two alternative interpretations are thus implied; (i) the interpretation of non-layered gabbros as "non-cumulate" suggests that similar textured allivalites must also be interpreted in terms of nucleation and growth, not crystal settling; (ii) it can also be argued that the gabbros have in fact formed by cumulus processes but for some reason do not display "igneous layering". This problem is discussed in Chapter 2.

Classification of the gabbros according to the scheme outlined in fig 3.5, is summarized in Table 3.1. There is a distinct correlation between texture and the form of the intrusion (and therefore the rate of consolidation); the veins have granular and discrete pyroxenes, whilst the large plug and sheet intrusions have predominantly subophitic and ophitic textures. Attention is also drawn to the correlation between pyroxene habit and olivine-content; in the olivine-rich gabbros the pyroxenes are subophitic or ophitic, in the olivine-poor or olivine-free hypersthene gabbros they are granular or discrete. This major change may be related to the cessation of olivine

crystallization by the peritectic reaction Fo + L = Cpx + Opx, and may be due to an important change in the kinetics of clinopyroxene crystallization at that point. Alternatively it may simply be a function of the composition of the liquid and the degree of compositional supersaturation that occurs under varying circumstances. Similar observations by Walker (1969) and Olmsted (1979) support the conclusion from this study that the change in habit of clinopyroxene is linked to the cessation of olivine crystallization.

3.4.3 Modal variation

Changes in the proportions of olivine, plagioclase and pyroxene are illustrated in fig 3.8 and Table 3.2. A representative suite of gabbro samples was point-counted and plotted in terms of the three major components. Ortho- and clinopyroxene were summed together, the minor amounts of opaques included with pyroxene, and alteration products, if present, were counted as the original primary minerals. The data illustrate the wide modal variation of the gabbros. The majority are olivine-bearing, and of these approximately half contain orthopyroxene. Olivine and plagioclase range 0-40% and 30-75% respectively. Pyroxene in the majority of gabbros is the dominant mafic phase present; Ca-rich pyroxene varies 4-48% and Ca-poor 0-20%. Intra-intrusion variations are considerable.

Data from gabbros of SW Rhum (Wadsworth, 1961) overlap with those of the Barkeval area (fig 3.8) and extend the spread of data towards the plagioclase apex. Some gabbros overlap with the plag-cpx-ol cumulates of the Dornabac Member and grade into the E.L.S. allivalites with increasing feldspar- and olivine-contents, while others, with increasing olivine-content, overlap with the Harris Bay Member of ol-plag-cpx cumulates (fig 3.8). Fig 3.8 also emphasizes the relationship between texture and mineralogy (3.3.2) in that textural

subdivisions are represented by well-defined clusters in modal composition, e.g. discrete and granular textures are most common in gabbros with little or no olivine (<15%).

3.4.4 Classification and description

Forty-five thin sections from a representative suite of gabbro plugs, sheets and veins have been examined; the gabbros are divided into six main groups.

3.4.4.1 Group 1: These gabbros occur outside the ultrabasic complex, but within the Main Ring Fault, in the Loch Gaimlich-Long Loch area (Map 1). Whilst several have been mapped by Dunham (1962, 1965) as "early" gabbros (samples EG, J50, E87, M36 - Map 2), the remainder (samples T4, T5, T5X, T12, M56) occur as small isolated exposures immediately adjacent to the Long Loch Fault, and are mapped for the first time. The Group 1 gabbros are very coarse-grained, show the occasional development of small (0.2 m) pegmatitic patches, and are noticeably chloritized and epidotized in hand-specimen; having a greenish hue.

The dominant mineral assemblage is olivine, clinopyroxene and feldspar (Table 3.1). Olivine (<6%) forms anhedral to subpoikilitic grains, is heavily serpentinized, and partly or completely converted to serpentine, chlorite and magnetite. Clinopyroxene, the dominant mafic mineral, is pale brown in colour, and varies from large (3-6 mm) ophitic and subophitic crystals (T4, E87) to clusters of 0.2-0.8 mm granular grains (T12, M56). Schillerization is rare and alteration varies from occasional blebs of brown amphibole to marginal or complete replacement by magnetite, green amphibole and chlorite (EG, T5). A few crystals show patchy extinction and bent cleavage, indicative of slight post-crystallization plastic deformation. Orthopyroxene occurs rarely as small grains (<0.3 mm) adjacent to olivine (T4, T5, T5X).

Plagioclase is very variable in grain-size (4 x 0.5 mm - 0.2 x 0.6 mm) and texture (laths and fine-grained mosaics of subrounded grains). A notable feature of Group 1 gabbros is the development, between mafic clumps, of spherical aggregates (1-2 mm diameter) of radiating feldspar laths (fig 3.9). Zoning of the plagioclase is strong (both normal and oscillatory) and the majority shows a slight clouding by very fine, oriented, exsolved ?iron-oxide particles. Bent albite- and pericline-twin lamellae, corrugated grain boundaries, fine subgrains adjacent to grain boundaries, and fine-grained polyhedral aggregates are the result of plastic deformation followed by high-temperature diffusion-controlled recovery polygonization and primary recrystallization processes (Hobbs *et al.*, 1976; Nicolas and Poirer, 1976). Since these textures are best developed near the Long Loch Fault and in the margin of the layered complex, it is suggested that fault movements and the emplacement of hot ultrabasic rocks generated the heat necessary for the initiation of the recovery processes. The plagioclase is often heavily altered and products include epidote, sericite, zeolites and calcite.

The opaque oxides (5-17 modal %) are notably skeletal and consist of ilmenite and titanomagnetite. Primary ilmenite occurs as homogeneous, only slightly oxidized, anhedral grains interstitial to the silicates; the titanomagnetite has been oxidized at low to moderate pressure and above 600°C, with the direct formation of "exsolved" ilmenite (Lindsley, 1962). Three textural forms of ilmenite - titanomagnetite intergrowth are present (Buddington and Lindsley, 1964): (a) fine "trellis" ilmenite lamellae in all $\{_{111}\}$ planes of the host spinel; (b) "sandwich" lamellae (25-35 μm) in one set of {111} planes - one or two lamellae per grain; (c) euhedral to anhedral inclusions of ilmenite, partly or totally included ("external

composite" and "internal composite" forms respectively). All three types can occur in one grain, and "exsolved" ilmenite contents vary from 1 or 2% —>60%. The ratio of primary ilmenite to Fe-Ti spinel can only be estimated, due to difficulty in distinguishing primary from exsolved ilmenite; the origin of all composite and sandwich ilmenite by advanced oxidation is questioned on textural grounds (Haggerty, 1976 p. Hg 9).

Accessory minerals include sphene, apatite and biotite, the latter being associated with the Fe-Ti oxides and showing alteration to chlorite. One sample (T5) contains scarce interstitial quartz.

In conclusion, the Group 1 "early" olivine gabbros are characterized by:

(a) ubiquitous extensive hydrothermal alteration

(b) radiate plagioclase texture

(c) skeletal or extreme interstitial development of the Fe-Ti oxides.

3.4.4.2 Group 2: The thin coarse ol-px-plag veins which are intimately associated with the Eastern Layered Series, and which have been termed "contemporaneous ultrabasic veins" (Brown, 1956), form the second group of gabbros. Olivine forms clusters of subhedral equant grains between the feldspar and pyroxene, whilst a few small ($\frac{1}{2}$ mm) grains are poikilitically enclosed by the latter. Large (3-5 mm) green clinopyroxenes form "discrete" crystals with subophitic margins, and the feldspars form an interlocking mosaic of large (2 x 0.5 mm), slightly zoned and strained, laths. Opaque minerals are rare. The Group 2 veins are very similar to the gabbros of Group 4.

3.4.4.3 Group 3: Group 3 gabbros postdate the layered ultrabasic rocks, and include the two large heterogeneous plugs on Barkeval (section 3.3). They are typically fresh, and olivine- and orthopyroxene-

bearing. Olivines in the homogeneous grey facies are large (3-4 mm), anhedral and contain magnetite along cracks and margins. In contrast, olivines of the picritic facies are smaller (2 mm), equant, lack the magnetite inclusions, and occasionally contain deformation lamellae. In both facies they are partly, or entirely, mantled by orthopyroxene reaction rims. The clinopyroxene is typically pale brown, subophitic to interstitial, and strongly schillerized at grain margins. The olivine-rich facies contains an additional green, non-schillerized variety, which poikilitically encloses olivines (0.3 mm) and is subophitic towards feldspar. Twinning, patchy extinction and corrugated grain boundaries are common, and alteration is limited to occasional intragrain blebs of brown amphibole. Subordinate orthopyroxene occurs adjacent to olivine (and often recrystallized) and as larger subophitic patches (2 mm diameter) independent of olivine.

Plagioclase laths (6 x 2 mm - 1 x 0.3 mm) show strong normal zoning from indistinct cores, and heavy patchy "clouding" (responsible for the characteristically dark weathered surfaces). As with Group 1 gabbros, the feldspars are often broken, and show bent twin lamellae and very patchy extinction; features indicative of deformation. Subsequent heating above the temperature of deformation resulted in extensive recrystallization; the temperature increase was also responsible for clouding of the feldspars and clinopyroxenes (Poldervaart and Gilkey, 1954; Carstens, 1975). Two possible causes of this metamorphism are: (a) localized heating resulting from intrusion of fine-grained basic material (dykes and veins); (b) reheating caused by intrusion of the unlayered peridotite mass of western Barkeval (Ruinsival Member of the Central Series). (b) is preferred since the ubiquity of clouding and recrystallization precludes localized heating.

Minor amounts (<6 modal %) of Fe-Ti oxides are present. The ratio of primary ilmenite to titanomagnetite (approximately 6:1) is greater than in Group 1 gabbros, and the ilmenite forms numerous discrete grains independent of, or adjacent to, the spinel. Both the primary titanomagnetite and the secondary granules (associated with alteration of the mafic phases) are densely clouded with "oxidation-exsolved" trellis ilmenite. "Sandwich" and "composite" inclusions are rare. Accessory brown biotite flakes consistently rim the Fe-Ti oxides.

Gabbro which intensely veins the layered allivalites and peridotites on Barkeval's summit plateau (samples E77, B64, E81, B75) is petrographically very similar to that of the plugs described above, except for its higher mafics-content, slightly larger ratio of orthopyroxene to olivine, and more granular texture of the pyroxenes.

In summary the olivine-hypersthene gabbros of Group 3 are characterized by:

(a) intense clouding of the feldspar and schillerization of the clinopyroxene;

(b) extensive deformation and thermal metamorphism textures;

(c) a high primary ilmenite to titanomagnetite ratio.

3.4.4.4 Group 4: The coarse-grained pyroxene-rich gabbros of Group 4 occur sparingly as small plugs and veins in the Dornabac and Long Loch Members of the Central Series, NE of Loch an Dornabac. Although they lack igneous layering, they texturally resemble the plag-cpx-ol cumulates of the Dornabac Member (figs 3.6 and 3.7).

Minor amounts of small (<0.5 mm) anhedral olivines are enclosed by randomly oriented plagioclase crystals (0.5 x 0.2 mm - 2 x 1 mm). The latter are weakly zoned, unclouded, but incipiently sericitized. As in Groups 1 and 3 grain boundaries are serrated, and laths often

broken and annealed. Clinopyroxenes (2-5 mm) confer a porphyritic texture to the gabbro, and show patchy extinction, twinning and incipient alteration to blebs of reddish-brown amphibole. Opaque minerals are rare (<0.5%) and include magnetite and occasional composite sulphide blebs (chalcopyrite and bornite).

In more olivine-rich samples (B6, B1) the clinopyroxene (6-8 mm) is poikilitic or subpoikilitic towards olivine. Opaque minerals are more abundant and include, in order of abundance, Cr-spinels enclosed in olivine, Cr-spinels between plagioclase laths, and small interstitial grains of homogeneous ilmenite and magnetite.

The textural similarity of Group 4 olivine gabbros, and their close spatial relationship,to Dornabac Member cumulates suggest they represent crystallization products of basic magma (containing a small percentage of plagioclase, clinopyroxene and olivine phenocrysts) isolated from the main magma body during a period of tectonic disturbance.

3.4.4.5 Group 5: Group 5 gabbros have limited distribution, only occurring as local concentrations of thin (5-10 cm wide) veins in Unit 8 of the E.L.S. on the northern slopes of Barkeval (near Triangular Loch), and in the Ruinsival Member of the C.S. Olivine is subordinate (<6%) and forms 1-2 mm anhedral crystals heavily altered to serpentine and magnetite. Clinopyroxene (48 modal %) forms large, brown, equant, subhedral discrete grains (2-6 mm); the majority are schillerized and heavily altered to amphibole and magnetite. Large crystals of plagioclase (2 x 1 mm - 0.5 x 0.2 mm) typically show deformation features, possibly due to movement during consolidation.

These veins contain the largest concentrations of opaque oxides seen in the Rhum gabbros (10-16 modal %): ilmenite is the sole primary

Fe-Ti oxide present, and forms large interstitial grains which partly enclose plagioclase and clinopyroxene. Reduction has resulted in the subsolidus "exsolution" of Fe-Ti spinel lamellae oriented along the {0001} rhombohedral planes of the host ilmenite. Other grains exsolve irregular blebs of ?rutile. The veins are also intersected by shear zones up to 0.5 mm wide.

In conclusion the olivine gabbros of Group 5 are:

(a) rich in discrete clinopyroxene grains;

(b) unusually rich in opaque oxides;

(c) have ilmenite as the sole primary Fe-Ti oxide, and which exsolves titanomagnetite.

3.4.4.6 Group 6: The homogeneous, even-grained, smooth-weathering gabbros of Group 6 are the freshest and finest-grained on Rhum. They are confined to the L.U.C., particularly its eastern half. Within the study area they occur as sheets cutting the heterogeneous gabbro (Group 3) in Atlantic Corrie, and as an enveloping sheath around a large Lewisian block south of Priomh Lochs (Map 1).

The dominant assemblage is cpx-opx-plag. Modal proportions are constant, and in the majority of cases variation in grain-size is minimal. Both clino- and orthopyroxene are characteristically granular in texture, occurring together in clusters of round equant grains (<0.25 mm). Pale brown clinopyroxene is dominant and often clouded with coarsely-exsolved magnetite. The plagioclase laths (0.5 x 0.2 mm) are weakly zoned, and lack the deformation features seen in the Group 1 and 3 gabbros. Fine-grained (0.2-0.1 mm) granular Fe-Ti oxides are uniformly disseminated throughout. One sample (GH2) contains occasional serpentine-chlorite aggregates which may be olivine pseudomorphs.

In summary the Group 6 hypersthene gabbros are: (a) very fine-grained; (b) granular, and (c) lack sign of deformation or thermal metamorphism.

3.4.4.7 Miscellaneous gabbros: While most gabbros in the An Dornabac-Barkeval area can be classified into one of six distinct groups, several display the characteristics of two or more groups.

(A) A large irregular mass of medium-grained gabbro (B7) which cuts the peridotite "tongue" due east of Long Loch (Map 2) was correlated by Dunham and Emeleus (1967) with the fine-grained olivine gabbro sheets and ring-dyke of eastern Hallival (belonging to Group 6 - see section 3.5). However, it contains abundant clusters of small (0.5 mm) equant olivines unusually rich in exsolved iron-oxide rods: - a texture atypical of Group 6 gabbros. Clinopyroxene texture, and the development of small patches of fine radiating plagioclase laths, are Group 1 characteristics; however, the dominance of ilmenite among the Fe-Ti oxides links it to the Group 6 gabbros.

(B) As described in section 3.2 the gabbro in the floor of Atlantic Corrie (B84, B88) is texturally similar to some allivalites in the E.L.S., although no modal layering is found. The olivines (av. 2 x 1 mm) are occasionally rimmed by orthopyroxene: a feature not found in the ultrabasic rocks. Several features distinguish this gabbro from the Group 3 gabbro in the cliffs above:(i) the plagioclase is unclouded and undeformed; (ii) the clinopyroxene is ophitic and subpoikilitic; (iii) primary ilmenite is less abundant, and (iv) numerous interstitial composite sulphide blebs (pyrrhotite, pentlandite and chalcopyrite) are present. Therefore it is either a separate intrusion or a local facies within the large heterogeneous Atlantic Corrie gabbro mass. Inadequate exposure precludes confirmation of either case.

3.4.4.8 Gabbro pegmatites: Several white zoned sheets, up to 20 cm wide, cut the northern tip of the Loch Duncan peridotite "tongue" (sample J17, Map 2). The central zones consist of discrete and subophitic clinopyroxene (av. 1 mm), zoned and sericitized plagioclase (av. 1 x 0.5 mm), and abundant euhedral apatite, sphene and ilmenite. The finer-grained, more pyroxene-rich margins contain less Fe-Ti oxides, apatite and sphene (suggesting fractionation by crystallization from the walls in). The sheets show incipient epidotization and chloritization.

Similar gabbroic sheets have intruded and altered the Harris Bay Member cumulates in the Glen Duain river section (samples M22, M24). Here too there is abrupt facies change from marginal white microgabbro to coarser-grained ("pegmatitic") zone: the microgabbro may represent the chilled margins of volatile-rich basic magma sheets containing plagioclase and clinopyroxene phenocrysts, whilst the axial zones resulted from the concentration of P, Ti, H_2O, K and Na.

Similar gabbro-pegmatite patches occur in ultrabasic rocks some 300 m south of Salisbury's Dam. At this locality (36359960) Torridonian arkose is seen above unchilled and unlayered peridotite; the sharp contact dips 45° south. The arkose immediately above the contact (sample M62) exhibits extensive partial melting, whilst the peridotite is extensively serpentinized and unusually rich in primary hydrous phases (brown amphibole and mica) and acicular apatite (see Chapter 5). Pale green coarse-grained feldspathic segregations up to 0.6 m across, occur in the peridotite immediately below the contact (sample M43). They, like the zoned sheets, consist of plagioclase laths (2 x 5 mm), subophitic clinopyroxene, interstitial amphibole, mica, antiperthitic alkali feldspar, and apatite, sphene and ilmenite. Alteration increases towards the centres of the segregations, again suggesting concentration of volatiles.

It is suggested that a hot H_2O-rich olivine mush was intruded into the Salisbury's Dam area: since only the upper contact is exposed the geometry of the intrusion is a matter for speculation. Heat loss through the roof extensively melted the overlying arkose: accumulation of volatiles beneath the roof led to formation of the pegmatitic lenses, which are therefore interpreted as residues from fractional crystallization of the picritic magma.

The occurrence, 50-100 m further north along the Kilmory River, of thin sheets of similar and even more fractionated material, indicates that such residues were occasionally filter-pressed to form transgressive sheets. The most evolved have syenitic compositions. These heavily weathered, medium to coarse-grained leucocratic sheets and dykes consist predominantly of oligoclase laths (4 x 2 - 2 x 2 mm) which are zoned to albite, show faint albite twinning, and are heavily sericitized at their margins. Interstitial areas consist of highly-irregular, heavily-altered, 0.2-0.5 mm crystals of orthoclase and albite, together with a wide variety of accessory minerals (<3-15 modal %), (i) amphibole present as euhedral flakes, pleochroic in pinkish-red to honey tints and often zoned to colourless or pale green rims (richerite); (ii) ilmenite, the sole opaque oxide, in the form of disseminated elongate flakes; (iii) euhedral apatite grains often enclosed by ilmenite or amphibole; (iv) euhedral to subhedral sphene (pleochroic colourless to pink) as single grains or in aggregates with flakes of (v) honey to red-brown pleochroic mica; (vi) rare zoisite and epidote; (vii) rare grains of aegerine-augite and riebeckite.

3.4.5 Discussion

Comparison of the Barkeval gabbros with those outside the study area suggest that the Barkeval "eucrite" and marginal gabbro (Brown,

1956) and the An Dornabac gabbro (Wadsworth, 1961) fall into the Group 3 category, and that the fine-grained olivine gabbro and Askival Plateau gabbro (Brown, 1956) belong to Group 6 and 5 respectively. Descriptions of the Glen Harris, Ruinsival and Papadil intrusions (Wadsworth, 1961) are inadequate for classification.

The observation that each group of gabbros is quite distinct suggests that all of the gabbro intrusions of any one group are contemporaneous. However, lack of cross-cutting relationships between the various gabbro bodies leaves their relative chronology indeterminable by direct means. By indirect means the following sequence is suggested: Group 1, 2, 3, 4, 5 and 6.

(1) The "early" gabbros of Group 1 are the oldest known basic igneous rocks on Rhum, intruded at a high level. (Hydrothermal alteration occurred as a result of fluids circulating at the time of intrusion, or set up by the intrusion, or set up by subsequent intrusions).

(2) The Group 2 gabbros, from their field relations, textures and mineral chemistry, are very closely related and intimately associated with the formation of the Eastern Layered Series, and therefore probably contemporaneous.

(3) The large heterogeneous plugs and veins of Group 3 brecciate and hence postdate the Eastern Layered Series. Additional age constraints are imposed by the fact that they predate the Ruinsival Member of the Central Series.

(4) The Group 4 gabbros, like those of Group 2, are closely related in time and space to the (younger) ultrabasic Dornabac Member (see Chapter 2).

(5) The Group 5 gabbros cut the younger Ruinsival Member.

(6) Youngest of all the Rhum gabbros are probably those of Group 6. This is implied indirectly from their very fine-grained nature, and lack of deformation and metamorphism, suggesting that they were intruded late into the relatively cold country rocks.

Correlation of those gabbros outside the study area is hampered by lack of detailed petrographic descriptions.

The above sequence indicates several distinct phases of gabbro emplacement throughout the period of accumulation of the L.U.C., each phase occurring within a particular portion of the complex. Brown (1956) linked the gabbros to later disturbance of the E.L.S., followed by uplift of the whole ultrabasic mass along the Inner Ring Fault ("lubricated" by the marginal gabbro). Wadsworth (1961) suggested that gabbro intrusion accompanied differential uplift of the two halves of the complex along the Long Loch Fault. According to Dunham and Emeleus (1967) intrusion of fine-grained olivine gabbro was associated with slight subsidence of the central ultrabasic block. If (a) the sequence 1-6 is correct, (b) the marginal gabbro did act as "lubricant" during uplift of the Eastern and Western Layered Series (Brown, 1956), and (c) the marginal gabbro does indeed belong to Group 3, then the intrusion of later gabbros (Groups 4-6) and formation of their "host" layered ultrabasic rocks (Dornabac and Ruinsival Members) continued *after* emplacement of the E.L.S. and W.L.S. This uplift (emplacement) in the time interval separating gabbro Groups 1-3 and 4-6 corresponds to the major break in the stratigraphy of the ultrabasic complex, described in Chapters 2 + 7.

3.5 Mineral Chemistry

Electron microprobe data for the olivines, pyroxenes, feldspars and Fe-Ti oxides from a representative suite of Barkeval-An Dornabac area gabbros are presented in Appendix A. Previous determinations of mineral compositions were by wet chemical and optical methods (Brown, 1956; Hughes, 1960b; Wadsworth, 1961). A total of 25 polished sections from Groups 1, 2, 3, 4 and 5 have been examined. Details of analytical techniques are given in Appendix A.

3.5.1 Olivine

Olivines range in composition from $Fo_{70.5}$ to $Fo_{78.5}$ (fig 3.10) and show a maximum intergrain variation of 1-2 mol % Fo. No zoning has been detected. In fig 3.10 analyses are compared with those from other basic and ultrabasic rocks on Rhum, and other gabbroic intrusions. Olivines from gabbros in the western half of the complex (Wadsworth, 1961) are more forsteritic and exhibit a smaller compositional range than olivines in the Barkeval gabbros. However, this may be a function of the method of determination, since Nwe (1976) has shown that olivine compositions obtained by electron microprobe are always less magnesian than those determined optically. Compositions slightly overlap with, and are continuous from, the most iron-rich analyses in the Rhum layered ultrabasic sequence. The range in composition for the Rhum gabbros is smaller, and generally more forsteritic, than that for the gabbros of Skye (Weedon, 1961), Ardnamurchan (Gribble, 1974) and Slieve Gullion (Gamble, 1979), but resembles the earliest-known olivines of Skaergaard (Nwe, 1976), Kaerven (Wager and Brown, 1967) and Kap Edvard Holm (Abbott and Deer, 1972). Roeder and Emslie (1970) concluded that the Mg/Fe^{2+} ratio in olivine is independent of pressure, temperature and other elements in the liquid, and dependent only on the Mg/Fe^{2+} ratio of the liquid from which it

crystallized. The liquid from which the Rhum gabbros crystallized was therefore more magnesian than liquids present in the other British Tertiary igneous centres.

CaO-content of the olivines ranges from 0.02-0.11 wt% and shows no appreciable change with fractionation (fig 3.11). The maximum variation within a sample is 0.05 wt%, and whilst in some cases the olivine rims are poorer in CaO than the cores (by ca. 0.02 wt%), the majority of grains show no Ca-zonation. Olivines in the L.U.C. have comparable CaO-contents, as do olivines in peridotite nodules from kimberlites, alkali olivine basalts, and alpine-type peridotites (fig 3.11). Data for olivines in large stratiform intrusions are scarce.

Natural and experimental data suggest that the CaO-content of olivine is a function of pressure (Simkin and Smith, 1970; Stormer, 1973), temperature (Ricker and Osborn, 1964; Watson, 1979) and CaO-content of the coexisting liquid (Watson, 1979). The Rhum olivines have contents typical of "plutonic" olivines (Simkin and Smith, 1970) but the magnitude of any pressure effect is unknown (Stormer, 1973). Contents are less than those predicted for olivine in equilibrium with basaltic liquids (0.16-0.3 wt% CaO for liquids containing 8-11 wt% CaO, Watson, 1979) and may reflect the easy diffusion of Ca out of the olivine lattice during slow cooling (Ferguson, 1978). CaO-contents of olivines will also be determined by the abundance and order of crystallization of other Ca-bearing phases. If olivine is the sole crystallizing mineral, Ca^{2+} (with its ionic radius similar to Fe^{2+}) would be expected to increase with iron-enrichment. This is not the case for the Rhum gabbros, and the low contents may, instead, be due to the cotectic crystallization of calcic plagioclase and olivine (Boone and Fernandez, 1971; Ferguson, 1978). Subsequent

diffusion of Ca out of olivine during slow cooling (Ferguson, 1978), and depletion due to serpentinization (Coleman and Keith, 1971) may contribute to the scatter of data in fig 3.11 both within and between samples, and account for the lack, and inconsistency, of CaO zoning.

MnO data are presented in fig 3.12. Contents vary from 0.25 wt% at $Fo_{78.5}$ to 0.50 wt% at Fo_{72}, showing a steady linear increase with iron-enrichment. Intergrain variation averages 0.05 wt% (maximum 0.07 wt%) and rims are generally 0.02 wt% richer in MnO than the cores. Large sample to sample differences in MnO-content (0.3 wt%) between olivines of similar Fo-content, are characteristic of the gabbros (fig 3.12) and may be a function of the order and relative abundance of crystallizing olivine, pyroxene and Fe-Ti oxides. Coprecipitation, and or subsolidus re-equilibration, of pyroxene and Fe-Ti oxides (both contain appreciable amounts of MnO) with olivine could produce the large range of MnO-contents for olivines of a given Fo-content. The relatively late-stage crystallization of Fe-Ti oxides, and the absence of large MnO-variation in samples lacking abundant ilmenite and magnetite (and clinopyroxene) suggests that subsolidus re-equilibration during slow cooling is the dominant factor.

MnO-contents slightly overlap with, and are continuous from, the contents for olivines from the L.U.C. (fig 3.12). Olivines of similar Fo-content in the Skaergaard (Nwe, 1976) and Thessalonika (Sapountzis, 1979) gabbros have lower MnO values (fig 3.12), possibly the result of crystallization from a MnO-depleted liquid.

NiO shows positive correlation with Fo, ranging from a maximum of 0.26 wt% at $Fo_{77.5}$ to a maximum of 0.125 wt% at $Fo_{70.5}$ (fig 3.13). The maximum within-sample variation is 0.02 wt%, and zonation of grains is not detected. For olivines of a given Fo-content the NiO-content can vary, from sample to sample, by as much as 0.1 wt%. Olivines from

the L.U.C. (fig 3.13) show a similar spread of NiO-contents. This scatter from the line of maximum NiO-content may be the result of one or more of the following factors: (a) variation of the olivine-liquid partition coefficient for Ni (D_{Ni}^{ol-liq}) with temperature (Hakli and Wright, 1967; Leeman, 1973; Leeman and Lindstrom, 1978), pressure (Mysen and Kushiro, 1979) and bulk composition of the liquid (Arndt, 1977; Hart and Davis, 1978); (b) fractionation of a sulphide liquid; and (c) post-crystallization metasomatic removal of Ni.

With regard to point (a), variation in confining pressure is negligible since all the liquids are assumed to have been retained during fractionation, within a single high-level magma chamber. The apparent temperature dependence of D_{Ni}^{ol-liq} is considered to be largely related to changing liquid composition (Arndt, 1977; Hart and Davis, 1978) and therefore the low and variable NiO-content of the olivines is, in all probability, the result of low Ni-contents of the silicate magma.

Considering factor (b), the very high distribution coefficient of Ni between sulphide and silicate liquids (approximately 275, Rajamani and Naldrett, 1978) will cause even trace amounts of sulphide fractionation to deplete the silicate liquid in Ni. Duke and Naldrett (1978) demonstrated that the Ni-content of olivine crystallizing from a liquid decreases rapidly when the liquid becomes saturated with respect to sulphur (i.e. an immiscible sulphide liquid is precipitated). Therefore, assuming the maximum NiO-contents are representative of sulphur-undersaturated compositions, and the olivines correspond to primary magmatic compositions, it can be concluded that the gabbro olivines crystallized from a sulphide-saturated and Ni-depleted silicate liquid (fig 3.14). The range of NiO values for a particular Fo-content can be accounted for by varying ratios of olivine to sulphide

being removed from the liquid (fig 3.14B, after Duke, 1979); supporting evidence for this conclusion is taken as the occurrence of "low-Ni" olivines in gabbros with abundant sulphides (e.g. samples B84, B75, E67).

Metasomatic removal of Ni (factor (c)) e.g. during serpentinization, is negligible since even the most heavily serpentinized olivines show "non-depleted" NiO values.

3.5.2 Pyroxenes

3.5.2.1 Major elements: Pyroxenes analysed from the gabbros are listed in Appendix A, and projected into the $Ca:Fe^{2+}:Mg$ system in fig 3.15. Neither Ca-rich nor Ca-poor pyroxenes show subsolidus exsolution lamellae and therefore all analyses represent solidus compositions (Brown, 1957; Nwe, 1976). The majority of clinopyroxenes are calcic augites; a few plot in the diopside, endiopside and salite fields (Poldervaart and Hess, 1951). Their compositions have a limited range from $Ca_{46.7}\ Fe_{11.2}\ Mg_{42.1}$ to $Ca_{43}\ Fe_{17}\ Mg_{40}$; those coexisting with orthopyroxene are slightly poorer in Ca. Variation within and between samples is small (<2.2 atomic %), and zonation is slight (maximum 1.6 atomic %). Whilst the majority of crystals show typical Fe-enrichment and Ca-depletion (with constant Mg), the "discrete" grains of Group 5 (E57, E82, B28) show reverse zonation. This is attributed to a sudden increase in f_{O_2} (Speidel and Osborn, 1967) which promotes the crystallization of Fe-Ti oxides (a high modal % is present in these samples) and thereby enriches the interstitial liquid in Ca relative to Fe.

The orthopyroxenes (bronzite and hypersthene) show a slight increase in Ca-content with fractionation ($Ca_{1.7}\ Fe_{22.6}\ Mg_{75.7}$ — $Ca_{4.2}\ Fe_{25.3}\ Mg_{70.5}$). Variation within and between samples is small, <2.1 atomic %.

Compositions of the Ca-rich pyroxenes overlap with those of the L.U.C. (fig 3.15), particularly with the allivalites ($Ca_{47.2}$ $Fe_{6.6}$ $Mg_{46.2}$ - $Ca_{42.1}$ $Fe_{10.1}$ $Mg_{47.8}$), as expected from their modal and textural similarities. The Ca-rich pyroxenes from the one-pyroxene gabbros lie on the magnesian-end of the trends for alkaline intrusions, e.g. Shiant Isles, whilst the clino- and orthopyroxenes from the two-pyroxene gabbros have similar compositions to those from the tholeiitic layered intrusions, e.g. Bushveld, Jimberlana, Kap Edvard Holm (fig 3.15).

3.5.2.2 Minor elements: Minor elements (Al, Cr, Mn, Ti, Ni were analysed) show considerable variation as a result of the changing bulk chemistry of the liquid, and the interplay of additional competing influences of temperature, chemistry of coexisting phases, and crystallization order (pressure and rate of cooling are assumed constant during growth of the pyroxenes).

Variation in Al_2O_3-content is shown in fig 3.16. For the orthopyroxene-bearing gabbros contents are relatively low (<3 wt%), consistent with crystallization from a tholeiitic liquid. Decreasing Al_2O_3 with fractionation (for the majority of clinopyroxenes) and the high $Al_2O_3{}^{cpx}/Al_2O_3{}^{opx}$ ratios (>1.0) are also characteristic of other tholeiitic intrusions (Brown, 1957; Brown and Vincent, 1963; Atkins, 1969; Campbell and Borley, 1974; Wilkinson, 1975; Nwe, 1975; Buchanan, 1979). However, the higher Al_2O_3 values of clinopyroxene in the one-pyroxene gabbros (3.0-4.6 wt%) are typical of clinopyroxenes from rocks of alkali basalt parentage (Murray, 1954; Wilkinson, 1956; Wilshire, 1967; Gibb, 1973). For both Ca-rich and Ca-poor pyroxene the maximum within-sample variation is 0.5 wt%. Zonation in the majority of grains involves core to rim depletion (maximum 2.0 wt%) irrespective of normal (decreasing) or reverse (increasing) Mg/Mg+Fe

zoning. Decrease in the Al_2O_3-content with fractionation may be due to (a) decreasing pressure, since Al^{3+} substitution into octahedral sites is greater at higher pressures (Yoder and Tilley, 1962; Verhoogen, 1962; Aoki, 1964), (b) decreasing temperature; this favours the replacement of Si in tetrahedral sites (Verhoogen, 1962), (c) increasing activity of SiO_2 in the liquid, since most of the Al in igneous pyroxenes is found in the tetrahedral position with Si (Kushiro, 1960; Le Bas, 1962), (d) decreasing activity of Al_2O_3 in the liquid (Segnit, 1953; Clark *et al.*, 1962), (e) increasing substitution of Cr^{3+}, Ti^{4+} or Fe^{3+} in the octahedral positions, providing charge balance for tetrahedral Al^{IV} (Campbell and Borley, 1974).

Factors (a) and (b) are negligible since pressure is assumed constant (all liquid being retained in a single high-level magma chamber) and temperature may have decreased only slightly (the gabbros being buffered by the large mass of still relatively hot ultrabasic rocks). Parameter (c) is not applicable since Al^{IV} content is constant with fractionation (fig 3.17). Therefore Al_2O_3-content of the liquid, and the nature of the ion(s) providing charge balance are the dominant controls on Al_2O_3-content of the pyroxenes.

Cr_2O_3-contents of the pyroxenes are given in fig 3.18. Cr^{3+}, with its high crystal field stabilization energy (CFSE), becomes concentrated in early-formed clinopyroxenes, and shows rapid exponential depletion with fractionation (0.95-0.03 wt%). The complete range of values can be found in a single sample, e.g. B84. The fractionation trend is continuous with that of the cumulus and intercumulus pyroxenes of the L.U.C.; the sharp drop in Cr_2O_3-content occurs in compositions intermediate between those of allivalite and gabbro (i.e. Mg/Mg+Fe 0.85-0.82). The orthopyroxenes have consistently low Cr_2O_3 values (<0.08 wt%). Intergrain variation differs widely from sample to sample

(0.02-0.6 wt%), irrespective of textural type. Zonal trends parallel the overall fractionation trend, with a maximum core to rim difference of 0.6 wt%. Reversed zonation (e.g. E87) is due to Cr^{3+} (in addition to Ti^{4+}) providing the charge balance for increasing Al^{IV}.

MnO-contents increase with Fe-enrichment, 0.05-0.28 wt% in the clinopyroxenes and 0.25-0.43 wt% in the orthopyroxenes (fig 3.19). Zonal trends in the majority of crystals parallel the fractionation trend, and core to rim differences range 0.02-0.14 wt%. However, in E82, E87, E57 (Group 5 gabbros) MnO decreases from core to rim (by 0.01-0.08 wt%) due to disequilibrium crystallization or depletion of MnO in the liquid by Fe-Ti oxide crystallization. Fig 3.19 also illustrates the concentration of Mn^{2+} preferrentially into the Ca-poor pyroxene, the result of its zero CFSE and ionic radius similar to that of Fe^{2+} (Campbell and Borley, 1974).

TiO_2-contents range from 0.5-1.9 wt% in the clinopyroxenes and 0.3-0.82 wt% in the orthopyroxenes; the former values overlapping with those of clinopyroxenes from the L.U.C. (fig 3.20). The majority of the Rhum diopside, salite and calcic augite is "titaniferous" (1-2 wt% TiO_2, Yagi and Onuma, 1967). Intergrain variation is small, maximum 0.2 wt%. Contents are higher than those of pyroxenes from the Jimberlana (0.26 - 0.6 wt%, Campbell and Borley, 1974), Salt Lick Creek (0.04-0.47 wt%, Wilkinson *et al.*, 1975) and Skaergaard (0.15-0.85 wt%, Nwe, 1975, 1976) stratiform intrusions, and comparable with values from the Lilloise (1.7-2.1 wt%, Brown, 1973) and Blue Mountains (0.7-2.5 wt%, Grapes, 1975) complexes of alkali basalt parentage.

Ti-trends of individual clinopyroxene grains involve core to rim increases of between 0.12 and 0.96 wt% (paralleling the overall fractionation trend, fig 3.20), and are due to increasing activity of TiO_2 in the liquid prior to the crystallization of ilmenite and titanomagnetite.

In addition to high activity of TiO_2, entry of Ti^{4+} into pyroxene (mainly in the octahedral sites) is favoured by:-

(a) high temperatures (Verhoogen, 1962);

(b) crystallization from SiO_2-undersaturated liquids (and hence high Al^{IV}-contents, Kushiro, 1960; Le Bas, 1962; Barberi *et al.*, 1971);

(c) crystallization from Cr^{3+}-depleted liquids, since with its zero CFSE Ti^{4+} cannot compete with Cr^{3+} for charge deficient octahedral sites;

(d) rapid cooling rates and rapid crystallization (Walker *et al.*, 1976; Shibata *et al.*, 1979) - but not applicable to the slowly-cooled Rhum gabbros.

Ti-Al relations (fig 3.21) reflect the spread in composition between Ca-rich and Ca-poor pyroxene; the former are able to accommodate more Ti and Al. The general spread along the Ti:Al = 1:4 line at low Al^{3+}-contents (<0.13 cations), i.e. more fractionated compositions (fig 3.16) implies substitution of the type $R^{+2}(Mg,Mn,Fe) + 2Si^{4+} \rightleftarrows (Ti^{4+} + Fe^{3+}) + Al^{+3}$ (Bence *et al.*, 1970; Grapes, 1975). The decrease in Ti/Al to 1/6 (for Al^{3+} >0.13 cations, i.e. more "primitive" compositions) implies the involvement of Cr^{3+} (fig 3.18) as the component $CaCrAlSiO_6$ (Nakamura and Coombs, 1973). Therefore with fractionation, the Ti/Al ratio increases from 1/6 to 1/4 as a result of decreasing activity of Cr^{3+} and Al^{3+} and increasing activity of Ti^{4+} in the liquid. Data presented in fig 3.21B show that pyroxenes from many alkaline and tholeiitic intrusions have Ti/Al ratios of approximately 1/4; those from Salt Lick Creek and Jimberlana have lower ratios (1/6) due to their higher Cr^{3+}-contents (fig 3.18).

Nickel in the pyroxenes (0-0.05 wt%) is just within the detection limit of the probe (see Appendix A) and minor amounts of Na_2O (0.30-

0.63 wt%) in clinopyroxene and 0.02-0.06 wt% in orthopyroxene) are normally present throughout the fractionation range.

3.5.2.3 Pyroxene as an indicator of magma type: Compositional trends (for both major and minor elements) of the Rhum gabbro pyroxenes may provide information on the genesis of the gabbros and the origin of the associated ultrabasic complex. Several workers have attempted to relate clinopyroxene chemical composition to the nature of the parental magma, i.e. tholeiitic or alkali basalt (Kushiro, 1960; Le Bas, 1962; Verhoogen, 1962; Coombs, 1963; Barberi *et al.*, 1971).

In terms of major and minor elements the Rhum pyroxenes appear transitional:

(a) the coexistence of Ca-rich and Ca-poor pyroxenes is typical of tholeiitic suites, but the more Ca-rich diopsides and salites of the one-pyroxene gabbros are common constituents of basic rocks of alkali olivine basalt parentage (fig 3.15). However, diopsidic-salitic pyroxenes have been reported in several subalkaline (tholeiitic) rocks (Norton and Clavan, 1959; Loeschke, 1973), and a one-pyroxene (Ca-rich)-bearing assemblage may be found in some rocks of tholeiitic parentage, due to changing phase relations with cooling, in the system Di-En-An (Lewis, 1973). Therefore considering major elements alone, the results are inconclusive.

(b) Si-Al^{IV} relations (fig 3.22) reflect the spread in composition between the Ca-rich and Ca-poor pyroxenes; the former contain more Al^{IV}. Data for the two-pyroxene gabbros spread across the tholeiitic field; clinopyroxenes from the one-pyroxene gabbros straddle the alkaline-subalkaline transitional zone and have Al^{IV}-contents typical of some alkaline complexes. They may, however, have anomalously high Al^{IV} due to a higher crystallization temperature (Verhoogen, 1962), or inaccuracies in the SiO_2 analyses. Fast cooling rates and increased

pressure, which favour entry of Al into the pyroxene lattice (Lofgren *et al.*, 1974; Shibata *et al.*, 1979) are not applicable to these coarse-grained gabbros.

(c) the low TiO_2-content (<0.8 wt%) of clinopyroxene in the orthopyroxene-bearing gabbros is consistent with their tholeiitic nature, whilst the 1-2 wt% TiO_2 in the pyroxenes of the orthopyroxene-free samples is suggestive of alkaline affinities (since high Ti^{4+} is favoured by high Al^{IV} and therefore low SiO_2-activity, Verhoogen, 1962). TiO_2-Al^{IV} relations (fig 3.23) show the spread of data across all compositional fields, although the majority plot close to the subalkaline-transitional boundary. Within-crystal zonation can extend plots into the alkaline field.

In summary, the clinopyroxenes have chemical characteristics similar to those typical of both tholeiitic and alkaline basalts (figs 3.15, 3.22, 3.23), suggesting crystallization from a liquid with physio-chemical properties transitional between the tholeiitic and alkali basalt series. However, caution is required when using pyroxene composition alone to interpret the host magma composition, since (i) figs 3.22 and 3.23 illustrate how the degree of fractionation (as typified by core to rim zoning) can seriously alter interpretation; (ii) factors other than the bulk rock composition can influence the clinopyroxene chemistry, e.g. temperature, pressure, rate of cooling (Kushiro, 1960; Verhoogen, 1962, Walker *et al.*, 1976; Shibata *et al.*, 1979); (iii) in some rocks variation in core compositions can exceed that of the core to rim (?due to differential clinopyroxene growth); (iv) the chemistry of the clinopyroxenes can be accounted for by a crystallization history which effectively masked differences due to initial liquid composition.

It is therefore concluded that the bulk rock chemistry may be a more reliable magma-type guide (see Chapter 6).

3.5.3 Plagioclase

Feldspar analyses are listed in Appendix A and shown in figs 3.24 and 3.25. Compositions of cores show a wide range from An_{85}-An_{50} and marginal zoning extends the range to An_{14}. The composition of the gabbroic feldspars overlaps those of the L.U.C. (An_{92}-An_{74}; zoned to An_{53}) and extends to distinctly more sodic compositions, suggesting a continuum between the two major groups (already concluded on textural and petrological grounds - sections 3.4.2, 3.4.3, 3.4.4).

With the exception of Group 6, plagioclases have been analysed from all the main gabbro groups, and the intergrain and intragrain compositional variations summarized in fig 3.25. The feldspars from Group 3 are labradoritic, with a restricted range An_{52}-An_{57}; marginal zoning is weak, e.g. $An_{53.6}$-$An_{52.8}$. In all other groups the plagioclase is more calcic. As is to be expected, those gabbros which are closest in texture to the allivalites contain the most calcic grains (An_{83}-An_{80}; zoned to An_{69}). Many gabbros contain a few small, indistinct cores of An_{85}-An_{84} which probably represent the original "phenocrysts" or "primocrysts", but are predominantly composed of labradorite, An_{68}-An_{56}. Typically the crystals show narrow, sharply defined outer zones with compositions extending to An_{44}. Exceptionally strong zoning to An_{14} is found in an "early" Group 1 gabbro (E87).

Similar feldspar compositions have been recorded from other Rhum gabbroic rocks (Brown, 1956; Wadsworth, 1961) as shown in fig 3.25; they were determined optically.

Several minor elements were analysed in the plagioclases; FeO ranges 0.30-0.74 wt%; MgO, 0.03-0.10 wt%; K_2O, 0.06-0.27 wt% and TiO_2, 0.03-0.11 wt%, and their distribution is controlled by a number of

factors. TiO_2 and K_2O increase and FeO and MgO decrease with Na-enrichment; the absolute variation is greatest for K_2O and FeO (0.19 and 0.41 wt% respectively), and least for MgO (0.01 wt%). These patterns are a reflection of the relative ease with which the minor elements are incorporated into the feldspar structure, and the changing composition of the melt during crystallization. The decreasing Fe-content with fractionation may indicate that the Fe/Na ratio in the liquid decreased with fractionation, either due to decreasing iron-content of the melt (as a result of the crystallization of the mafic phases) or due to slower iron-enrichment than sodium-enrichment. Alternatively f_{O_2} may have controlled the Fe-trend. Assuming (a) the majority of the iron in the plagioclase is ferric (replacing Al^{3+}), and (b) the Fe^{2+}/Fe^{3+} ratio of the feldspar reflects the Fe^{2+}/Fe^{3+} ratio of the liquid, then the decreasing Fe-contents may reflect progressively more reducing conditions (decreasing f_{O_2}) in the liquid during fractional crystallization. The increase in TiO_2-content from core to rims is taken to reflect the smooth continuous increase in TiO_2-activity in the liquid, prior to the crystallization of the Fe-Ti oxides.

3.5.4 Iron-titanium oxides

With the exception of Group 2 and 4, all the gabbros contain Fe-Ti oxides in the form of interstitial titanomagnetite grains with trellis, sandwich and composite ilmenite intergrowths (section 3.4.4.1), together with homogeneous ilmenite grains in contact with, or independent of, titanomagnetite. Modal proportions of the opaque oxides are given in Table 3.2. In all gabbros they have clearly crystallized at a late stage. Analyses from the complex intergrowths are listed in Table 3.3 and Appendix A.

Wide variations in composition of the oxidation assemblages which developed from the primary Fe-Ti spinel and ilmenite, are a function of (a) the composition of the initial liquid (and hence the composition of the initial Fe-Ti oxides), (b) the degree to which oxidation has proceeded, and (c) the absolute values of temperature and f_{O_2} during the crystallization history.

3.5.4.1 Ilmenite: The term "ilmenite" refers to an ilmenite-haematite solid solution. Ilmenite which crystallized originally as discrete grains is referred to as "primary"; the lamellae and laths of trellis, sandwich and composite ilmenite are termed "secondary" or "exsolved".

Ilmenite analyses are plotted in fig 3.26 in terms of wt% proportions of TiO_2 ($+SiO_2$), RO (FeO + MnO + MgO) and R_2O_3 (Fe_2O_3 + Al_2O_3 + Cr_2O_3). Total iron-contents (determined as ferrous, by the probe) have been recalculated into FeO and Fe_2O_3 using the proceedure given by Carmichael (1967); mol % endmember compositions calculated according to Carmichael (1967) are listed in Table 3.3. Recorded CaO, Na_2O and SiO_2 values are due to silicate contamination. Compositions range from $Il_{99}\ Hm_1$ - $Il_{90}\ Hm_{10}$ and are comparable to those from other basic intrusives. There are no systematic differences in Il-Hm composition between similar textural types from the main gabbro groups. Although the majority of the ilmenite, both primary and exsolved, is homogeneous and unoxidized, some (e.g. B75, B7) shows slight oxidation: the development of a small number of tapering needles (2-5μm) of rutile throughout the grains (characteristic of the R_2 stage of oxidation, Haggerty, 1976). Assuming no loss or gain of Ti, oxidation processes will take place along Ti/Ti + Fe isopleths (fig 3.26) such that late-stage oxidation of an original composition A will give a final bulk composition B, consisting of a ferrian ilmenite

host C and exsolved rutile D. Individual components are too fine for analysis. However the bulk composition B (obtained by the probe) can be reduced at constant Ti/Ti + Fe to give the original primary composition A.

Several minor elements (Mn, Al, Cr, Mg, Ni) were also analysed. Contents of Cr_2O_3, NiO and Al_2O_3 are low and range 0.01-0.33 wt%, 0.01-0.11 wt% and 0.04-1.57 wt% respectively. MgO is present as the geikilite component ($MgTiO_3$); contents (0.14-5.75 wt%) are plotted in fig 3.27 and they overlap with the field of primary intercumulus ilmenites from the allivalites and to a lesser extent with the primary and composite-type exsolved ilmenite in the cumulus Cr-spinels of the layered peridotites. Fields of ilmenite from the Kap Edvard Holm, Skaergaard, Somerset Dam and Bushveld intrusions are also shown for comparison.

The MgO-content of ilmenite, previously thought to be pressure influenced (cf. the high contents so characteristic of kimberlites) is now believed to be primarily a function of the composition of the parent magma (Haggerty, 1976 p.Hg 155). Since olivine is the dominant MgO-bearing phase to crystallize prior to ilmenite, the MgO-content of the initial magma, and the degree of olivine fractionation are constraints on Mg-entry into ilmenite. This is evident from fig 3.27 which indicates a correlation between ol-content of the rock and Mg-content of the ilmenite present. An additional factor is the partitioning behaviour of Mg^{2+} between ilmenite and magnetite: Mg preferentially partitions into ilmenite, primary or exsolved (Table 3.3 and Stormer, 1972).

In summary, the geikilite concentration in ilmenite is a function of (a) MgO-content of the initial magma, (b) the degree of fractionation of ferromagnesian minerals, and (c) the crystallization sequence.

Contributory factors include (d) the temperature of ilmenite formation, (e) the rates and paths of equilibration; ilmenite continually equilibrates with coexisting silicates and therefore changes composition with cooling (Usselman, 1975).

Manganese, the other minor element present in substantial amounts, ranges from 0.34-2.56 wt% MnO (fig 3.28); contents increase with decreasing temperature (Buddington and Lindsley, 1964; Lipman, 1971; Neumann, 1974) and MnO is strongly partitioned towards the ilmenite regardless of whether it be primary or exsolved (Czamanske and Mihalik, 1972; Neumann, 1974).

Since on textural grounds distinction between primary and oxidation-exsolved ilmenite is often difficult (section 3.3.4.1) many workers have considered mineral chemistry as a useful indicator of ilmenite type. Evans and Moore (1968), Lipman (1971), Duchesne (1972) and Mathieson (1975) noted differences in MnO; Fe, Ti, Mg and Mn; Ti and mol % Hm; and Ti,Mn,Fe^{3+}-contents respectively. Differences are apparent in the Rhum ilmenites, and similar to those reported by Mathieson (1975) from the Somerset Dam layered basic intrusion: primary ilmenite has higher recalculated Fe^{3+} and mol % Hm, higher wt% Cr_2O_3 and slightly higher Al_2O_3 than exsolved ilmenite, whilst the latter has higher FeO, MnO, TiO_2 and slightly higher MgO.

Differences between the various textural types of secondary ilmenite are less consistent, and much less marked. This is to be expected since (a) the Il-Hm contours of the Buddington and Lindsley geothermometer (1964) are approximately parallel to the buffer lines, and (b) it is generally accepted that the cooling curve of a magma, in the f_{O_2}-T field, normally parallels the buffer lines. Therefore with cooling, successive ilmenite generations will be similar in composition. The greatest change will be shown by the titanomagnetite.

3.5.4.2 Titanomagnetite: The term titanomagnetite refers to members of the solid solution series between ulvospinel and magnetite. Analyses are listed in Table 3.3 and plotted in terms of wt% TiO_2 + SiO_2, RO and R_2O_3 components in fig 3.26. Fe_2O_3-contents are calculated according to the method of Carmichael (1967), and mol % Usp-Mt compositions according to Powell and Powell (1977).

Compositions range from Usp_4Mt_{96} to $Usp_{23.3}Mt_{76.7}$; they do not represent primary magmatic compositions since the titanomagnetite has been oxidized to produce trellis ± sandwich and composite ilmenite. Oxidation has induced a characteristic partitioning of the minor elements: MgO (0.08-3.66 wt%) partitions preferentially into the ilmenite, Al_2O_3 (0.72-5.95 wt%), Cr_2O_3 (0.14-13.13 wt%) and MnO (0.02-0.5 wt%, fig 3.28) favour the spinel structure. Variation in TiO_2 (1.09-6.8 wt%) is a function of TiO_2-levels in the primary spinel, and the degree of "contamination" of the microprobe spot by trellis ilmenite. CaO and SiO_2 are assigned to silicate contamination (Vincent *et al.*, 1957). Since, within any one sample, all magnetite grains contain very fine trellis ilmenite, it is not possible to compare the compositions of magnetite which coexists with trellis only, sandwich only and composite ilmenite only.

Fig 3.29 shows analyses plotted in terms of Al^{3+}, Cr^{3+} and Fe^{3+} in the ternary projection of the multicomponent spinel prism, together with titaniferous spinels from the ultrabasic rocks. There appears to be a continuous spinel series from chromites within olivine in the peridotites and allivalites, through the disseminated spinels of the allivalites, to interstitial titanomagnetites of the gabbros. However, in zoned spinels, the contacts between chromite cores and titaniferous magnetite rims are relatively sharp (figs 3.30 and 3.31). These discontinuities are most likely the result of a peritectic

involving the precipitation of pyroxene in the interval between chromite and titanomagnetite crystallization (Irvine, 1965, 1967): the absence of spinel in rocks containing cumulus pyroxene (Chapter 2), and experimental data on the compositions of spinels in basalts crystallized over a range of f_{O_2}'s and temperatures (Hill and Roeder, 1974) confirm this.

The intermediate compositions (i.e. between chromite and titanomagnetite) may be primary (Thompson, 1973) or secondary, the result of reaction of early-formed Cr-rich spinels with residual liquid during the end stages of crystallization (Evans and Moore, 1968; Gunn *et al.*, 1970; Evans and Wright, 1972; Ridley, 1974, 1977). The disseminated spinels from the allivalites have reacted with trapped liquid: lack of change in Cr/Al ratios from cores to rims precludes reaction with cumulus plagioclase (Chapter 2 and Henderson, 1975).

3.5.4.3 Ilmenite-titanomagnetite geothermometry and oxygen geobarometry: The experimental results of Buddington and Lindsley (1964) enable the equilibration temperature and oxygen-fugacity of coexisting magnetite and ilmenite to be determined from their compositions. Compositions of fast-quenched Fe-Ti oxides of volcanic rocks give liquidus temperatures and oxygen-fugacities (Carmichael, 1967; Anderson, 1968a); data from slowly-cooled rocks are more difficult to interpret because of subsolidus re-equilibration (Anderson, 1968a; Duchesne, 1972; Mathieson, 1975; Bowles, 1976, 1977; Oliver, 1978; Rollinson, 1979).

Electron microprobe analyses from complex ilmenite-titanomagnetite intergrowths are presented in Table 3.3. Mol % Usp-Mt and Il-Hm values, and the re-allocation of total iron (as FeO) into FeO and Fe_2O_3 can be calculated in several ways, to minimize the as yet unknown effects of

minor amounts of RO and R_2O_3 components. The various methods are reviewed by Bowles (1977): that of Carmichael (1967) is adopted in this study. Due to oxidation of some original Fe^{2+} to Fe^{3+}, the total FeO-contents of the titanomagnetite are too low. Estimates of Usp were therefore obtained using the relation wt% $TiO_2/35.75$ (Powell and Powell, 1977). Since the trellis ilmenite lamellae in the spinels are too fine to be analysed separately, a defocused beam was used, and bulk spinel compositions obtained.

Temperatures and oxygen-fugacities obtained from the Buddington and Lindsley (1964) calibration curves, for the recalculated present compositions of sandwich and composite ilmenite and titanomagnetite (including trellis ilmenite), are listed in Table 3.3 and presented in fig 3.32A. The temperatures (540-600°C) and f_{O_2}'s ($10^{-23.5}$-$10^{-19.3}$ atm.) define a curve parallel to, and approximately coincident with the QFM buffer curve. Allowing for uncertainties in the experimental data of the f_{O_2}-T curves, values are within ±30°C and $\pm10^{-1}$ atm. for coexisting pairs (Buddington and Lindsley, 1964). The temperatures (540-600°C), obviously too low for magmatic crystallization, reflect "blocking" temperatures below which diffusion could not continue to form the sandwich and composite ilmenite: they are a function of the rate of diffusion and amount of the exsolved phase present (Rollinson, 1979). Subsequent cooling, and the implied continuous oxidation and Ti-diffusion on a smaller scale, produced the final trellis generation of ilmenite. Final equilibration temperatures and f_{O_2}'s could not be determined due to the difficulty in obtaining trellis analyses. Lamellae of spinel, exsolved from ilmenite (samples E57 and E82) plot on the reducing side of the QFM buffer curve.

Ilmenite-titanomagnetite pairs from different grains within the same sample, have equilibrated at different T and f_{O_2} conditions

(subject to the limits of error given above), suggesting the rocks contained several independently equilibrating closed systems. Core to rim primary or secondary (resulting from limited diffusion during homogenization) zonation has not been studied in detail.

Original high-temperature Fe-Ti oxide compositions can be deduced by several methods (Mathieson, 1975; Bowles, 1977): that of Mathieson (1975) is used here. Several assumptions are necessary:

(a) in compliance with the Buddington and Lindsley (1964) hypothesis, all exsolved ilmenite is assumed to result from the oxidation of Usp in an original Usp-Mt solid solution;

(b) primary ilmenite and magnetite have crystallized contemporaneously, and in equilibrium with each other and the melt;

(c) effects of minor element contents are minimal (see recalculation method above).

By combining the present compositions of magnetite and exsolved sandwich and composite ilmenite, in the proportions given by point count modal %'s in the aggregates, the original magnetite compositions can be estimated. Pre-oxidation compositions of primary ilmenite grains (distinguished by their chemistry, section 3.5.4.1) are deduced by reduction along the Ti/Ti + Fe isopleths to the Il-Hm_{ss} join (fig 3.26). They are taken to represent original magmatic compositions, since the existence of compositional differences between primary and secondary ilmenite indicates that the primary grains did not apparently undergo full sub-solidus re-equilibration.

The inferred original T and f_{O_2}'s (Table 3.4 and fig 3.32A) plot close to the QFM buffer curve, and reflect the conditions in the intermediate to late stages of crystallization. The Group 6 gabbro

data (e. g. E57), plot on the reducing side of the QFM buffer as the result of loss of the buffering phase (?H_2O). H_2O may have been consumed during uralitization of the pyroxenes (a petrographic characteristic of this group, see section 3.3.4.6). That buffering was controlled by an H_2O-rich fluid, is also suggested by the intimate growth relationship between Fe-Ti oxides and biotite ± amphibole. Similar conclusions were reached by Vincent and Phillips (1954), Elsdon (1972) and Parsons (1980). Silicate phases have not controlled the subsolidus f_{O_2}, since cooling curves are similar for rocks of widely differing silicate modal proportions.

T-f_{O_2} data from other gabbroic intrusions, are illustrated in fig 3.32B. Before significant conclusions can be drawn, the method of recalculation must be considered: widely differing results are obtained from the various methods (Bowles, 1977). However, analyses recalculated by the methods of Anderson (1968a) and Carmichael (1967) differ by only 13°C and $10^{-0.5}$ atm. (well within the limits of experimental error), and therefore the Rhum Fe-Ti oxides can be considered to have equilibrated at temperatures similar to those of the Somerset Dam and Kap Edvard Holm intrusions. Dissimilarity of the Skaergaard data is attributable to (a) different calculation method, and (b) lower f_{O_2} and water-content in the Skaergaard magma.

3.6 Summary

1. Four mineral assemblages are present:
   - (i) plag, cpx;
   - (ii) plag, cpx, ol;
   - (iii) plag, cpx, opx;
   - (iv) plag, cpx, opx, ol.

   Types (ii) and (iv) are most abundant.

2. Gabbro textures (based on clinopyroxene habit) show complete gradation between discrete, ophitic, intergranular and granular types.
3. The change from ophitic to granular clinopyroxene habit is linked with cessation of olivine crystallization.
4. "Non-cumulate" gabbro textures grade into "cumulate" allivalite textures.
5. Six petrographically distinct gabbro-types are present: Groups 1-6.
6. The suggested sequence of intrusion (oldest-youngest) is: Group 1, 2, 3, 4, 5, 6.
7. Groups 1-3 were intruded prior to and during emplacement of the L.U.C.: Groups 4-6 post-date the emplacement.
8. Olivine compositions range from $Fo_{78.5}$-$Fo_{70.5}$: they are "Ni-depleted" due to sulphide fractionation.
9. Clinopyroxenes are Ti- and Al-rich calcic augite, diopside and salite: orthopyroxenes are bronzite and hypersthene.
10. Plagioclase compositions range An_{85}-An_{50}, extended to An_{14} by marginal zoning.
11. Equilibrium Fe-Ti oxide pairs (ilmenite-titanomagnetite) show subsolidus re-equilibration down to 540-600°C and f_{O_2}'s of $10^{-23.5}$-$10^{-19.3}$ atm: buffering was controlled by an H_2O-rich fluid.

CHAPTER 4

THE DYKES

4.1 Introduction

Prior to the nineteen-seventies little work had been done on the fine-grained basaltic minor intrusions of Rhum. Harker (1908) noted the marked variation in their distribution between the various country rocks, and concluded they were (a) most abundant in the Torridonian strata, (b) relatively rare in the Western Granophyre, (c) concentrated in the eastern portion of the ultrabasic complex, and (d) belonged to a general regional swarm trending NW-SE, but (e) showed a marked radiate pattern and increased frequency towards the igneous complex. Richey (1961), Stewart (1965) and Dunham and Emeleus (1967) also refer to the existence of a radial dyke swarm on Rhum.

Speight (1972), as part of a regional study of dyke distribution patterns in NW Scotland, re-examined the Rhum swarm. His study of over 1000 dykes along the NW and SE coastal sections confirmed the existence of a main linear NW-SE swarm, and a smaller NE-SW "subswarm" within the Torridonian strata, but failed to support the concept of a radial swarm. The possibility of a radial swarm within the ultrabasic complex was suggested by Forster (1980).

Truncation of dykes at major contacts implies several major episodes of dyke intrusion, e.g. dykes in the felsites do not cut the adjacent gabbros (Hughes, 1960b; Dunham and Emeleus, 1967), and dykes in the Western Granophyre are truncated by later gabbro and peridotite (Wadsworth, 1961). Recently Forster (1980) has made a major contribution to knowledge of the sequence and timing of the dyke intrusion episodes. By detailed mapping along well exposed contacts, and interpretation of Rose diagrams, he concluded that (a) the Main Linear Dyke Swarm, MLDS (Speight, 1972), itself produced by several intrusive

events, is truncated by the Main Ring Fault, and truncated and thermally metamorphosed by mafic and ultramafic rocks on Minishal, (b) rarity of fine-grained basic blocks in the explosion breccias suggests the majority of the dykes within the Main Ring Fault post-date the breccias, (c) rare occurrences of composite dykes indicate contemporaneous acid-basic magmatism (e.g. Stony Corrie, Dibidil), (d) most dykes within the Main Ring Fault post-date the emplacement of the acid rocks and pre-date emplacement of the ultrabasic complex, and (e) several dykes cut the lavas and therefore represent the youngest known magmatic events on Rhum.

Forster (1980) presents the first petrographic and geochemical data on the dykes. The work presented here (Chapters 4 and 6) supplements his study by concentrating on those dykes within the ultrabasic complex.

4.2 Field Relationships

The term "dyke", as used in this study, is defined as an intrusive discordant sheet-like body which can vary in attitude from vertical to a dip of 65°. If less than 0.1 m wide the term "vein" is used. Sills, inclined sheets and parallel inclined sheets (defined, Forster, 1980) are rare.

4.2.1 Field characteristics

The dykes are small (<0.75 m wide and mostly 0.1-0.4 m) and rarely traceable for over 20 m. Thickness is generally constant, although the finer-grained dykes have very tortuous paths in which they repeatedly thicken and thin, terminate sharply and continue en echelon along strike. Tachylitic margins are rare: in most cases the dykes show only a slight fining of grain-size towards margins. One dyke (B65) shows a coarse-grained spinifex-texture (Nesbitt, 1971)

with dendritic olivines ranging from 1-4 cm. Three major groups of dykes (and veins) are found:

Group A: have olivine and spinel phenocrysts. Olivine phenocryst-contents are extremely variable (<5-60 modal %), but an average content is estimated at approximately 20%. Because of their olivine-rich nature, the group A dykes are termed "picrites" (Drever and Johnston, 1967). Groundmasses are medium-grained: in some cases they are fine-grained and quenched. Weathered surfaces are characteristically brown.

Group B: are extremely fine-grained and grey weathering. They are aphyric or mildly phyric (<5 modal %), containing phenocrysts of olivine, feldspar and clinopyroxene. They are basaltic.

Group C: are aphyric, extremely fine-grained and leucocratic (with <5% mafic minerals), with a characteristic sucrose texture, and will be referred to as aplites.

4.2.2 Trends and distribution

Trends of dykes recorded from eight parts of the study area are presented as Rose diagrams in fig 4.1. Areas of high dyke-density correspond with areas of intense normal faulting: dykes parallel the faults. Three dominant fault/dyke directions are seen: (a) a major set trending NW-SE, (b) a major set NE-SW, and (c) a small number trending N-S. The dyke directions cluster around each major trend, forming three groups 35°-50°, 350-020° and 305-320° (fig 4.1). The Main Linear Dyke Swarm consists of three similar components (NW-SE and NE-SW swarm and subswarm, Speight, 1972, and a N-S subswarm, Forster, 1980), which suggests a radial fracture system did not develop above the Rhum magma chamber. The three fracture directions are parallel to pre-Tertiary lines of crustal weakness (a) the NE-SW Caledonian "grain", (b) the NW-SE trending horsts and grabens of

Triassic and Jurassic age in the North Sea Basin (Zeigler, 1975), and (c) N-S "geofractures" initiated in the early Carboniferous (Russell, 1971). This also suggests a radial fracture system did not develop: the magmas utilized pre-existing crustal fractures.

4.2.3 Age relations

Group B dykes are the most common: they also intrude the gabbro plugs and vein stockworks (Chapter 3), and the small peridotitic minor intrusions (Chapter 5). Cross-cutting relationships indicate that at least two intrusive events are represented. Several dykes cut the fine-grained marginal gabbro in Coire Dubh, thereby implying that at least part, if not all, of the Group B injection episode took place after emplacement of the ultrabasic complex.

Group A picrites occur predominantly on the western and southern flanks of Barkeval, where they post-date the heterogeneous gabbro plugs (Chapter 3). Detailed mapping shows that several ages of picrite are present. Group B dykes pre- and post-date the picrites, and a third generation of picrites cuts the Ruinsival Member of the Central Series. Isolated picritic dykes are also found in the Salisbury's Dam area, the Ard Mheall Member, Coire Dubh, and the Minishal extension of the Long Loch Member. A fourth generation may be present, within the Main Linear Dyke Swarm; they have been truncated and thermally metamorphosed by the Minishal peridotite (Forster, 1980), and therefore pre-date the emplacement of, at least, this Minishal portion of the Long Loch Member (Chapter 2).

The Group C aplites are rare, found only at two localities (B37 and B86, Map 2). Both localities (in the heterogeneous gabbro plugs) consist of local concentrations of 2-6 cm wide sub-parallel veins with sharp, straight contacts. Veins can be traced for up to 10 m before they pinch out. Their age relative to Groups A and B is unknown.

4.3 Petrography

Within each of the three major dyke groups (A-C) considerable petrographic variation is found, and division into subgroups is based on (a) the characteristic phenocryst assemblages and contents, and (b) the mineral assemblage and textures in the groundmass. A representative suite of rocks has been examined; modal analyses and phenocryst assemblages are given in Table 4.1. The majority of samples are very fresh.

4.3.1 Group A

Olivine and minor Al-Cr-spinel are the sole phenocryst phases. The dykes are variably porphyritic, with olivine phenocrysts contents varying from <5-60%. Groundmasses are equally variable, ranging from medium-grained ophitic and subophitic to fine-grained quench textures. Three subgroups have been recognized:

A-1: Olivine phenocrysts (10-40 modal %) occur singly or in clusters, and range from euhedral to rounded subhedral, equant, and from 0.5-5 mm. The majority show marginal recrystallization and, or, reaction rims in the form of fine-grained mosaics of olivine and amphibole. The olivines are fresh and dusted with iron-oxide.

Spinels occur as large (0.6 mm) single homogeneous anhedral phenocrysts (2-3 modal %), and as homogeneous subhedral grains (0.1-0.3 mm) partially or totally enclosed within olivine phenocrysts.

The groundmass consists of olivine grains (0.15 mm) and zoned plagioclase laths (0.1 x 0.5 - 0.2 x 0.7 mm) poikilitically and ophitically enclosed by brown schillerized clinopyroxene, and variable amounts of strongly pleochroic red-brown to honey coloured amphibole. Interstitial opaque phases comprise abundant irregular blebs of Fe-Cu-Ni sulphides (chalcopyrite, pyrrhotite and pentlandite) and Fe-Ti oxides (magnetite and ilmenite): the latter often enclose the former.

Abundance of primary amphibole (and in some samples, mica) is a characteristic feature of A-1 picrites, and indicates that liquid differentiated towards a Ti-rich, undersaturated residual fraction. The presence of numerous sulphide blebs indicatesthat the liquid was sulphur-saturated just before or immediately after emplacement, and the presence of abundant groundmass olivine and brown (?titaniferous) clinopyroxene implies alkalic affinities.

Three dykes from Salisbury's Dam (M57, M63) and Coire Dubh (M39) show considerable hydrothermal alteration: the olivine, pyroxene and amphibole are partially converted to serpentine, chlorite, carbonate and magnetite, and the plagioclase is saussuritized. M63 contains rare large (7x2 mm) lath-shaped plagioclase phenocrysts.

A-2: Members of this subgroup cut (and are occasionally cut by) the A-1 picrites; some post-date the Group B basalts. They lack primary amphibole (and mica) but otherwise are petrographically identical to the A-1 picrites. Occasional specimens contain rare flakes of a pleochroic lime-green to colourless amphibole (?pargasite).

One specimen (E53) is extremely fine-grained, showing quenched margins rich in skeletal olivine (extreme hopper and branching types -- fig 4.2) and a "flow" fabric defined by parallel alignment of H-shaped quenched plagioclase crystals. Olivines in the vein centre have non-skeletal morphologies (granular and polyhedral). The phenocryst content is ca. 15%. The crystallization of abundant skeletal olivines in the selvedges suggests that the liquid was picritic rather than basaltic, and the systematic change from margin to core, of hopper and branching to polyhedral and granular olivines, results from the decreasing degree of supercooling and rate of crystallization with cooling (Donaldson, 1976).

Another variant, sample B81, consists of sparse fresh subhedral olivine phenocrysts (single or in clusters, 0.5-2 mm, and ca. 5%)

and glomerophyric clusters (2-6 mm) of olivine and plagioclase, in a fresh medium-grained groundmass of granular olivine (0.2 mm) and plagioclase laths (0.2-0.4 mm) poikilitically and ophitically (respectively) enclosed by pale green clinopyroxene. Opaque phases are rare; a few small spinels are enclosed in olivine phenocrysts, and interstitial blebs of Fe-Cu-Ni sulphides are present in the groundmass.

A third variant, B65, is of petrographic importance. It is characterized by skeletal crystals (quench texture) and absence of phenocrysts, and probably represents an original liquid (Chapter 6). Delicate "hopper", "crystallographic-branching" and "linked parallel growth" olivine morphologies (Donaldson, 1976) are present, and crystals range from 3-20 mm in length. Orientation is random, and despite the occasional deformation lamellae (indicative of strain) the majority of crystals are unbroken. Plagioclase forms bundles of skeletal laths, intergrown with the olivines, and of equal dimensions. Laths are slightly strained (bent twin lamellae) and clouded. Interstices are filled with strongly zoned plagioclase, subophitic brown clinopyroxene, primary ilmenite and titanomagnetite (in approximate 1:1 ratio), and numerous composite sulphide blebs.

A-3: This small group of picrites has a very limited distribution, being found at only four localities (samples E71, J81, B9, M9, see Map 2). Each locality consists of a small concentration (2 x 2 m^2) of sinuous 10-20 cm wide veins and dykes of characteristically black-weathering picritic material - "black picrite". Chills and evidence for flowage differentiation are absent.

Olivine and spinel phenocrysts (55-65 and 2-4 modal % respectively) show a large and continuous range in size, <0.5-7 mm and 0.2-1 mm respectively. The olivines, irrespective of size, are predominantly euhedral to subhedral equant (polyhedral and granular, Donaldson, 1976). A few large grains are "hopper" type. All show intense clouding by

very finely exsolved "trails" of iron oxide, as a result of high-temperature oxidation (Haggerty and Baker, 1967a), and hence their characteristic black weathered appearance. A variant of this clouding is found in M9, where the olivines are brown-coloured. The olivines show variable degrees of marginal alteration to serpentine, chlorite and magnetite (maximum in J81, minimal in M9) as a result of reaction with interstitial, and or, hydrothermal liquid (Haggerty and Baker, 1967b). Some contain deformation lamellae.

Microphenocrysts of homogeneous euhedral and subhedral translucent (brown) and opaque (black) spinels are relatively common (2-4 modal %) and those in quenched groundmasses are zoned. Contacts between the two spinel "species" can be sharp or gradational. Similar zonal patterns are common in spinel phenocrysts from all basalt types (Evans and Moore, 1968; Evans and Wright, 1972; Thompson, 1973; Ridley, 1977). Smaller crystals (unzoned) are commonly included in the olivine phenocrysts and are surrounded by a 4-5 μm wide zone of clear olivine, the result of subsolidus re-equilibration between spinel and oxidized olivine.

The groundmasses are generally medium-grained (av. 1 mm) and ophitic, and comprise clinopyroxene, plagioclase, Fe-Ti oxides and minor amounts of amphibole and mica. The brown clinopyroxene has undulatory extinction and shows marginal alteration to a pleochroic blue-green amphibole. Plagioclase forms clumps of sub-parallel laths (2 x 0.2 mm), often with cloudy centres and marked continuous zoning. Some laths are strained, and patches are heavily altered to analcite and zeolites. The primary Fe-Ti oxides consist of interstitial grains of ilmenite and magnetite (the latter often with vermicular edges), whilst numerous specks of secondary magnetite are associated with oxidation and alteration of the olivine and pyroxene.

Several hydrous phases are present in the groundmass: (1) abundant red-brown to honey pleochroic amphibole (?kaersutite) zoned to pleochroic green amphibole (?pargasite) occurs as (a) interstitial flakes between and within plagioclase, (b) plates subpoikilitic towards olivine, and (c) rims to clinopyroxene. Textures (a) and (b) are suggestive of the amphibole having crystallized at a late stage from a volatile-rich residual liquid. Amphibole (c) may be primary overgrowth onto clinopyroxene, or secondary due to reaction of pyroxene with late interstitial liquid.

(2) flakes of pleochroic red-brown to honey coloured mica rim the Fe-Ti oxides and are enclosed by clinopyroxene and amphibole.

Some A-3 picrites are quenched (e.g. M9) with groundmasses consisting of sheaths of sub-parallel and partially radiating plagioclase laths (maximum length 1.5 mm and often hollow-cored and H-shaped) and interstitial clinopyroxene, magnetite and ilmenite (fig. 4.3).

The presence of minor interstitial blebs of sulphide in some samples indicate that these liquids were just sulphur-saturated at the time of emplacement.

4.3.2 Group B

The group B basalts cut, and are cut by, the A-1 and A-2 picrites. They are extremely fine-grained and aphyric or with scarce microphenocrysts of olivine, plagioclase and clinopyroxene. Phenocryst-contents are lower and the silicate assemblages more complex (ol-plag-cpx; plag-ol-cpx; plag-cpx; cpx-plag; and plag) than in the picrites (Table 4.1). The groundmasses are less variable in texture and composition and generally more leucocratic. Chilled margins are common.

Olivine phenocrysts (up to 4 mm) occur singly, in clusters, or in glomerophyric clumps with plagioclase (e.g. M45), and are equant euhedral (polyhedral) or rounded subhedral (granular). Hopper forms

are occasionally found. The degree of high-temperature oxidation is variable, alteration slight, and deformation lamellae are common in the more olivine-rich samples.

Rare spinel microphenocrysts (<1%) occur only in the more olivine-rich dykes. They are unzoned, up to 0.6 mm across, euhedral to subhedral and oxidized (trellis, sandwich and composite ilmenite exsolution). Rare grains enclosed in olivine are smaller (<0.1 mm), round and homogeneous.

Clinopyroxene phenocrysts (0.5-2 mm) are found in the majority of Group B dykes (Table 4.1) and occasionally in glomerophyric clusters with plagioclase and olivine (M45). All are pale green (some zoned to narrow pale brown rims, e.g. M193), fresh, rounded subhedral and equant.

Plagioclase is a ubiquitous phenocryst phase throughout the suite (<1-5 modal %). Crystals occur as single euhedral laths, or in glomerophyric clusters with olivine and pyroxene, and range in size from 0.3 x 1 mm to 2 x 4 mm; some are strained (curving twin lamellae). The majority are strongly zoned, with normal continuous or discontinuous zoned narrow overgrowths on subhedral,and often oscillatory zoned, cores.

Groundmasses are fine-grained (<0.3 mm), quenched, intergranular or granular in texture, and composed of plagioclase, clinopyroxene and opaques; olivine has been identified in only one sample (M193). Most consist of randomly oriented acicular H-shaped (quenched) plagioclase, with granular aggregates of pale brown clinopyroxene, Fe-Ti oxides (1:2 ratio of ilmenite and oxidized titanomagnetite) and accessory amounts of Fe-Ni-Cu sulphides (pentlandite, pyrrhotite and chalcopyrite). In the more leucocratic examples (M45, M197) the feathery plagioclase laths have a variolitic texture. Sample M45 is thermally metamorphosed

(cloudy feldspars, highly-serrated grain boundaries and polygonal strain-free aggregates) due to intrusion of the nearby Ruinsival Member.

4.3.3 Group C

The Group C aplites have syenitic compositions and consist of alkali-feldspar, Fe-Ti oxides ± quartz (Table 4.1). B37 consists of Carlsbad-twinned microperthite (braided type) and minor (<5%) disseminated aggregates of ilmenite and less magnetite. The aplitic veins have also been thermally metamorphosed by the Ruinsival Member. In contrast B86, a quartz syenite aplite, lacks a metamorphic texture, and consists of an equigranular mosaic of untwinned cryptoperthite, and micrographic alkali feldspar-quartz intergrowths (15-20 modal %), with disseminated Fe-Ti oxides (predominantly ilmenite) and rare aggregates of serpentine and magnetite (?originally pyroxene or amphibole).

These aplites are probably genetically related to the syenitic segregations of the Salisbury's Dam area (Chapter 3, section 3.4.4.8).

4.4 Mineral Chemistry

A total of 26 polished sections from the three groups (A-C) have been examined, and microprobe data for the olivines, spinels, clinopyroxenes, amphiboles, feldspars and Fe-Ti oxides, together with details of analytical techniques, are presented in Appendix A.

4.4.1 Olivine

Both phenocryst and groundmass olivines show a wide range in composition, $Fo_{92.5}$-$Fo_{72.5}$ and $Fo_{84.4}$-$Fo_{69.9}$ respectively (fig 4.4). Olivines in the picrites (Group A) span the complete range, with each subgroup forming a distinct cluster of data: $Fo_{83.8}$-$Fo_{79.9}$ (A-1), Fo_{77}-$Fo_{72.5}$ (A-2) and $Fo_{92.5}$-Fo_{85} (A-3). Group B consists of only 2

samples (since olivine is a rare phase) but shows a comparable range of compositions: Fo_{79} (phenocrysts in B2) to $Fo_{69.9}$ (groundmass in M193). The correlation of the trimodal distribution of olivine Fo-contents with distinct petrographic subgroups precludes the possibility that the pattern is a reflection of sampling bias. Consequently, the compositional gaps are considered real.

Comparisons of Fo-content with olivines from other basic and ultrabasic rocks on Rhum, are summarized below.

Dykes	$Fo_{92.5}$-$Fo_{72.5}$	(this study)
Ultrabasic Series	$Fo_{89.4}$-$Fo_{79.0}$	" "
Gabbros	$Fo_{78.5}$-$Fo_{70.5}$	" "
Peridotitic Minor Intrusions	$Fo_{91.1}$-$Fo_{80.0}$	" "
Lavas	$Fo_{85.0}$-$Fo_{50.0}$	(Emeleus, pers. comm.)

The maximum intergrain variation within any one sample is 0.35 mol% Fo. Zoning in the majority of olivines is negligible, but in the quenched dykes (e.g. B62/2, M9) zoning is normal and continuous, with a maximum core to rim decrease of 4 mol% Fo (av. 2.5 mol%).

Sample M9 is noteworthy since it shows a relationship between texture and chemistry of the olivines. (a) the strongly-coloured brown olivine phenocrysts, irrespective of size, have the highest Fo-contents ($Fo_{92.5}$-$Fo_{89.2}$ zoned to $Fo_{87.1}$) and are crowded with small rounded translucent brown spinels.

(b) the colourless olivine phenocrysts range from $Fo_{89.6}$-Fo_{88} (zoned to $Fo_{86.5}$) and contain opaque spinels in addition to the translucent variety.

(c) the sparse brown granular groundmass olivines are unzoned, lack spinel inclusions and are assumed to have quench compositions ($Fo_{84.4}$-Fo_{83}).

M9 is therefore characterized by a large variation in olivine composition, a high phenocryst-content (>55 modal %), a limited morphological range of olivines (polyhedral and granular, with occasional hopper-type; Donaldson, 1976), and a quenched groundmass. The presence of >55% phenocrysts implies the possibility of enrichment in cumulus olivine, but several lines of evidence argue against formation of the picrite by low-pressure cumulus addition of olivine to less magnesian associated liquids.

(1) the difference in phenocryst composition between M9 ($Fo_{92.5}$-Fo_{88}) and the A-1, A-2 picrites and Group B basaltic dykes (Fo_{79}-$Fo_{72.5}$) precludes the possibility that M9 has been derived from them by accumulation of olivine. This result is further substantiated by the difference in major element composition between the groundmass of M9 picrite and the whole rock composition of the aphyric Group B basaltic dykes (Chapter 6).

(2) Fe-Mg partitioning between olivine and liquid (Roeder and Emslie, 1970) precludes an accumulative origin for the M9 olivine phenocrysts: the calculated Fo-content of the first-formed olivine (Fo_{92}) is within 0.5 mol% Fo of the highest observed composition ($Fo_{92.5}$), and the K_D value (0.34) is within the range of generally accepted equilibrium values (0.30-0.34, Cawthorn *et al.*, 1974). Compositions of the M9 olivines ($Fo_{92.5-87.7}$) are within the range of assumed mantle olivine compositions (O'Hara, 1968) and indicate only limited fractionation of the M9 liquid.

Evidence therefore suggests that picritic (high-MgO) magmas were delivered from unknown depths to the high-level environment, without losing the crystals formed. The bulk compositional control of these magmas is represented (with regard to major elements) by an olivine fractionation curve (Chapter 6), and might imply that the magmas

(a) were already variably olivine-phyric when intruded, only some of the observed olivine having grown at low pressure, or (b) were delivered in a liquid state, having suffered olivine fractionation at some earlier stage. However (b) is not considered further since the lack of skeletal olivine and the wide range of Fo-contents exclude in-situ crystallization during quenching (the presence of olivine nuclei at the time of quenching explains the non-skeletal morphologies of the groundmass olivines, Donaldson, 1976).

The polyhedral and granular olivine morphologies for a wide range of Fo-contents, imply that for the magma (liquid + crystals) the degree of supercooling (low) and rate of cooling and crystallization were comparatively uniform over a considerable vertical distance. The mechanism termed "compensated crystal settling" (Cox and Bell, 1972; Krishnamurthy and Cox, 1977), may explain how the magma did not experience a net loss of crystals during rapid ascent.

In summary, M9 picrite is considered to be of great petrological importance. It is suggested that the brown olivines ($Fo_{92.5}$-$Fo_{89.2}$) crystallized at depth from a high-MgO liquid in equilibrium with mantle olivines (the chemistry of the enclosed translucent spinels also indicates a high-pressure origin - see section 4.4.4.1). As crystallization continued during rapid ascent, the lower temperature, lower pressure olivines and opaque spinels formed, until ultimate quenching at low pressure. The significance of such picritic magmas on Rhum is discussed in Chapter 6.

CaO-contents of the olivines (fig 4.4) range from 0.44-0.02 wt% (the majority of crystals containing 0.02-0.12%) and have a bimodal distribution (<0.2 wt% and >1.2 wt% CaO), with each data group showing no appreciable change with fractionation. Maximum intergrain variation is 0.08 wt% and generally less than 0.06 wt%. Rims are lower by an

average of 0.04% CaO, but maximum zonation is shown by the olivine (skeletal and non-skeletal) in quenched dykes. Within any one sample groundmass olivines have consistently higher CaO-contents than the phenocrysts.

Natural and experimental data suggest that the CaO-content of olivine is a function of pressure (Simkin and Smith, 1970; Stormer, 1973), temperature (Ricker and Osborn, 1964; Watson, 1979) and CaO-content of the coexisting liquid (Watson, 1979). Although the majority of olivines have low CaO-levels (<0.1 wt%) consistent with a "plutonic" environment (Simkin and Smith, 1970) the high CaO-contents of picrite M9 olivines are inconsistent with their deduced high-pressure origin. However, the M9 olivines have CaO-contents higher (0.2-0.44 wt%) than those predicted for olivines in equilibrium with a basaltic liquid (0.16-0.3 wt%, Watson, 1979) and may therefore have crystallized from a liquid more calcic than basalt (i.e. >11 wt% CaO). Although the estimated CaO-content of the M9 liquid (9.41 wt% - Chapter 6) is typically basaltic, it is significantly more calcic than liquids of similar MgO-content (Searle and Vokes, 1969; Clarke, 1970; Elthon and Ridley, 1979).

Skeletal olivines do have CaO-contents consistent with crystallization from a basaltic liquid, but olivines from the medium-grained (non-quenched) picritic and basaltic dykes are depleted in CaO. This may reflect the ability of Ca to diffuse out of the olivine lattice in the slower-cooled dykes (Ferguson, 1978); the fact that comparable CaO-contents (<0.1 wt%) are found in olivines from the slowly-cooled layered ultrabasic rocks and gabbros (fig 4.4B) is taken as supporting evidence for this conclusion.

It has been suggested that CaO-contents will be greatly determined by the order of olivine crystallization relative to other Ca-bearing

phases (Boone and Fernandez, 1971; Ferguson, 1978) and therefore higher CaO-levels should be found in olivines from the picrites (no plagioclase phenocrysts) than in olivines from glomerophyric clusters (assumed to have co-precipitated with feldspar). That this is not the case, is a reflection of the Ca-depletion of the picritic olivines due to diffusion during and following emplacement; the higher CaO-contents at some olivine rims may be attributed to such a diffusive mechanism.

MnO-contents of the olivines are presented in fig 4.5. Contents vary between 0.1 wt% at $Fo_{92.3}$ and 0.54 wt% at Fo_{70}, showing a steady linear increase with fractionation. Intergrain variation averages 0.05 wt% but can be as much as 0.2 wt%, and rims are generally 0.05 wt% richer in MnO than the cores. Within any one sample the groundmass olivines are richer in MnO than the phenocrysts, despite often similar MgO-contents. For a given Fo-content the dyke olivines are slightly richer in MnO than olivines from the layered peridotites and unlayered gabbros, but have comparable contents to olivines from the layered allivalites.

The NiO-content of the olivines decreases with decreasing Fo-content from a maximum of 0.49 wt% at $Fo_{92.3}$ to a maximum of 0.22 wt% at $Fo_{75.5}$ (fig 4.6). The intergrain variation is generally less than 0.03 wt% (maximum 0.12 wt%) and all grains show a decrease of NiO from core to rim (average 0.03 wt%, maximum 0.1 wt%). Sample to sample variation in NiO for olivines of a given Fo-content, can reach 0.18 wt%; olivines from the L.U.C. and the gabbros show a similar spread (fig 4.6).

Several factors may have produced this scatter from the "maximum-NiO" line.

(a) metasomatic removal of Ni during serpentinization: this is not considered further, since the degree of serpentinization does not correlate with Ni-content.

(b) decrease in D_{Ni}^{ol-liq} as a result of increased pressure (Mysen and Kushiro, 1979): with the exception of M9, all the dyke olivines are considered to have crystallized at low pressures, hence pressure dependence is negligible. In addition, the decrease in Ni-content with fractionation, in M9 (which crystallized under conditions of rapidly decreasing pressure) precludes the low Ni-contents being a result of decreases in D_{Ni}^{ol-liq} (since decreasing pressure would increase D_{Ni}^{ol-liq}; Mysen and Kushiro, 1979).

(c) decrease in D_{Ni}^{ol-liq} due to decrease in Ni-content of the magma (Clarke and O'Hara, 1979; Elthon and Ridley, 1979): theoretical data suggest that fractionation of Ni-sulphide will significantly deplete co-precipitating olivines in NiO (Duke and Naldrett, 1978; Duke, 1979). Natural data also suggest that separation of a sulphide liquid must play an important role, since many basalts are saturated or nearly saturated with sulphur at eruption (Skinner and Peck, 1969; Mathez, 1976), and many MgO-rich liquids have undergone Ni-sulphide fractionation (Naldrett and Cabri, 1976). The very high value of D_{Ni} between sulphide and silicate melt (ca. 275, Rajamani and Naldrett, 1978) indicates that even if only trace amounts of sulphides fractionate, D_{Ni}^{ol-liq} will have little significance. The "low-Ni" olivines in the Rhum dykes are therefore considered to have crystallized from a sulphur-saturated, Ni-depleted silicate liquid. Up to 3 modal % sulphides (pentlandite and pyrrhotite) are recorded.

Because of the dominant role of olivine in the fractional crystallization of basaltic liquids, the Ni-content of the olivine has been considered a sensitive petrogenetic indicator (O'Hara *et al.*, 1975; Sato, 1977; Hart and Davis, 1978; Clarke and O'Hara, 1979; Elthon and Ridley, 1979). However, in view of the above discussion, interpretation of Rhum Ni-partitioning data will only be significant when analyses

plotting on, or very close to, the line of "maximum NiO" are used. Nickel distribution between olivines and liquid is summarized in Table 4.2

Cr_2O_3-contents range from 0.15 wt% at $Fo_{92.5}$ to 0.08 wt% at Fo_{83}, showing a steady decrease with fractionation. In more iron-rich compositions Cr_2O_3 is below the detection limit of the microprobe (see Appendix A, Table A.2). Chromium is believed to enter into the olivine structure as Cr^{2+}, substituting for Fe^{2+} (Burns and Burns, 1975).

4.4.2 Feldspar

Feldspar analyses are listed in Appendix A and shown in fig 4.7. Groups A and B show similar wide ranges of groundmass plagioclase compositions (An_{76}-An_{46} and An_{73}-An_{43} respectively, the most calcic occurring in the A-3 picrites); strong marginal zoning extends the ranges to distinctly more sodic compositions (An_{33}). Plagioclase phenocrysts (absent in the Group A picrites) range An_{89}-An_{55} and are extended by marginal zoning to An_{43}. Orthoclase in the Group C syenitic aplites has a limited compositional range Or_{42}-Or_{55}.

The compositions of the dyke feldspars overlap those of the gabbros (An_{85-50} + zoning to An_{14}) and of the L.U.C. (An_{91-74} with zoning to An_{53})-see fig 3.24.

Several minor elements were analysed; FeO ranges 0.20-1.43 wt%; MgO, 0-0.53 wt%; and TiO_2, 0-0.17 wt%. Zonation is marked. The most calcic phenocrysts (An_{84}-An_{69}) show an increase in FeO (maximum 0.57 wt%), MgO (maximum 0.80 wt%) and TiO_2 (maximum 0.11 wt%) with increasing Na_2O-content, reflecting the increasing concentration of these elements in the liquid, prior to the crystallization of clinopyroxene and Fe-Ti oxides. In contrast, the more sodic phenocrysts (An_{66}-An_{60}) of the more evolved dykes show the same steady increase in TiO_2 (maximum core to rim difference 0.03 wt%) but exhibit decreasing FeO (0.78

wt%) and MgO (0.50 wt%); this is due to the appearance of TiO_2-poor clinopyroxene on the liquidus (these dykes, e.g. M47, M193, contain clinopyroxene microphenocrysts with <0.82 wt% TiO_2). All groundmass feldspars show core to rim zonation involving decreasing FeO, MgO and TiO_2 (maximum differences 0.57, 0.80 and 0.11 wt% respectively), as a result of their co-precipitation with Fe-Ti oxides.

4.4.3 Pyroxene

Phenocryst and groundmass pyroxene analyses are listed in Appendix A, and their compositional range shown in fig 4.8. Phenocrysts (Group B only) range from diopside ($Ca_{46.5}\ Mg_{43.8}\ Fe_{9.7}$) to calcic augite ($Ca_{40.6}\ Mg_{48.0}\ Fe_{11.4}$). Varying Ca/Mg ratios at constant Fe may be due to varying degrees of plagioclase and Al-bearing magnetite fractionation prior to clinopyroxene crystallization. Compositions (in terms of varying Fe^{2+} at constant Ca:Mg ratio) are transitional between those of the ultrabasic complex (Fe_{6-11} atomic %) and the gabbros (Fe_{11-17} atomic %). Phenocrysts from the Rhum lavas are distinctly more Fe-rich for a similar range in Ca:Mg ratios; Fe_{14-20} atomic %, Emeleus, pers. comm. (fig 4.8). Compositional variation within and between samples is limited (<2.0 Ca:Mg:Fe atomic %) but individual crystals show marked core to rim zoning involving Fe-enrichment with slightly decreasing Ca (maximum $Ca_{45.3}\ Mg_{45.3}\ Fe_{8.6} \rightarrow Ca_{40.4}\ Mg_{40.8}\ Fe_{18.8}$). Irrespective of Ca:Mg ratio, zoning parallels the fractionation trends for the Shiant Isles Sill (alkaline) and the Kap Edvard Holm intrusion (alkaline affinities)-see fig 4.8.

Groundmass pyroxenes (diopside, salite and calcic augite, fig 4.8) show a larger variation in Ca:Mg and Fe:Mg ratios. Those of Group B have a similar range of Ca (48-39 atomic %), but higher Fe^{2+}-contents (Fe_{14-21} atomic %) than the phenocrysts. The pyroxenes of Group A have the highest Ca-contents (44-49 atomic %), with each subgroup having a

distinct, mutually exclusive range of Fe^{2+} atomic % (Fe_{7-9} for A-1; Fe_{10-14} for A-2; Fe_{16-18} for A-3). Zoning involves core to rim increase in Mg with slightly decreasing Fe (Ca_{47} $Mg_{40.5}$ $Fe_{12.5}$ → $Ca_{42.5}$ $Mg_{46.5}$ $Fe_{11.0}$). This trend may be due to either a non-equilibrium (metastable) partitioning between crystals and liquid, or the presence of a zone of liquid depleted in pyroxene components. The former is preferred since Ti and Al are equal in quench and non-quenched pyroxenes. Groundmass pyroxene compositions in the very fine-grained dykes overlap considerably with those in the lavas, whilst compositions in the coarser-grained non-quenched picritic dykes are closer to those of the gabbros and ultrabasic complex (fig 4.8). This is clearly due to differences in cooling and crystallization rates, yet experimental studies have indicated no such variation in major elements as a function of cooling rate (Grove and Bence, 1977; Grove, 1978; Walker *et al.*, 1976; Coish and Taylor, 1979).

As a result of the incorporation of substantial quantities of Al, Ti, Cr, Mn, (Na + Ni) into their lattices, the pyroxenes show considerable variation in minor element chemistry.

MnO-contents increase with fractionation, 0.14-0.24 wt% in the phenocrysts, and 0.07-0.22 wt% and 0.21-0.39 wt% for the Group A and B groundmass grains respectively (fig 4.9). The lower rate of increase in MnO with Fe-enrichment for the picritic dykes, than for the Group B suite, may be due to a slower build-up of MnO in the liquid as the result of pyroxene crystallizing contemporaneously with abundant groundmass olivine (±Fe-Ti oxides). Zoning of phenocrysts is strong (maximum core-rim difference of 0.21 wt%) and parallel to the overall fractionation trend for the Group B suite. From fig 4.9 it should be noted that (a) the dykes (especially Group B) and gabbros have similar MnO vs Mg/Mg+Fe patterns, and (b) the phenocrysts have similar MnO-contents,

for a given Mg/Mg+Fe ratio, to the "discrete" pyroxenes of the gabbros and the cumulus pyroxenes of the allivalites.

Cr_2O_3-contents (fig 4.10) decrease exponentially with fractionation in both phenocrysts and groundmass (1.22 wt% at Mg/Mg+Fe 0.84 to 0.0 wt% at Mg/Mg+Fe 0.63). The Cr-rich phenocrysts (0.33-1.22 wt% Cr_2O_3) are strongly zoned parallel to the fractionation trend (0.70-<0.01 wt%). Groundmass pyroxenes in Group A have higher Cr_2O_3-contents (1.1 - 0.05 wt%) than crystals in Group B (0-0.18 wt%). Intergrain variation is maximum in the more magnesian compositions (0.4 wt%) and least in the iron-rich (0.04 wt%). The fields of pyroxenes from other lithologies are also shown on fig 4.10. As with the MnO diagram, the dyke and gabbro Cr_2O_3-fields are coincident, and the phenocrysts overlap with the cumulus pyroxenes in the layered allivalites and "discrete" (?cumulus) pyroxenes in the unlayered gabbros.

Titanium-contents (fig 4.11) range from 0.16-2.96 wt% and increase with fractionation. The phenocrysts are low in TiO_2 (0.67-1.05 wt%), with a maximum intergrain variation of 0.2 wt%. Individual crystals are moderately zoned (maximum core to rim increase of 0.2 wt%) but show a smaller rate of increase with Fe-enrichment than the overall fractionation trend. This may be due to their slower cooling rate (Coish and Taylor, 1979, Shibata *et al.*, 1979). TiO_2-contents are similar to those of "discrete" clinopyroxenes in the gabbros and cumulus grains in the allivalites.

Groundmass compositions span the complete range from low-TiO_2 (0.15 wt%) through "titaniferous" (1-2 wt%) to titanaugites and titansalites (2.0-2.96 wt%); groups A and B have similar contents (0.15-2.47 wt% and 0.43-2.5 wt% respectively). Within-sample variation is greatest in the phenocrysts (0.5-1.47 wt%), and within any one sample the groundmass grains have higher TiO_2-contents than the phenocrysts;

in keeping with their more Fe^{2+}-rich compositions and also due to their rapid cooling rate and crystallization (crystal growth is greater than the rate of diffusion of "impurities", such as Ti, away from the crystal; Walker *et al.*, 1976; Coish and Taylor, 1979; Shibata *et al.*, 1979). Higher contents in the more fractionated compositions imply increasing activity of TiO_2 in the liquid prior to the crystallization of Fe-Ti oxides.

Al_2O_3-contents (fig 4.12) vary from 2.2-4.86 wt% in the phenocrysts and from 1.25-7.34 wt% in the groundmass grains. Within-sample variation is generally large; 0.5-1.71 wt% (phenocrysts) and 0.1-3.73 wt% (groundmass). Zoning of the phenocrysts generally involves core to rim depletion (maximum 0.9 wt%), the reverse of the fractionation trend. Falling pressure and temperature are implied by the decreasing substitution of Al into the octahedral (VI) and tetrahedral (IV) sites respectively (Verhoogen, 1962; Aoki, 1964)-see fig 4.14. Increasing activity of SiO_2 in the liquid may also account for decreasing Al^{IV} (the bulk of the Al^{3+} in clinopyroxene, Kushiro, 1960; Le Bas, 1962) -see figs 4.13 and 4.14 - as may decreasing activity of Al_2O_3 in the liquid (Segnit, 1953; Clark *et al.*, 1962) and increasing substitution of Ti^{4+} (fig 4.15) in the octahedral site . The clinopyroxene phenocrysts are of low-pressure origin since their Al^{IV}:Al^{VI} ratios (pressure-controlled, Aoki and Shiba, 1973) are similar to those of the groundmass pyroxenes (av. 3:1 fig 4.13).

Groundmass pyroxenes, however, define a trend of increasing Al_2O_3 with Fe-enrichment (fig 4.12). Since a high proportion (75%) of the Al^{3+} is in the tetrahedral site with Si (figs 4.13 and 4.14), the increase in Al^{IV} is due to decreasing silica-activity in the liquid. This is characteristic of the fractionation of alkali basalt. Al_2O_3 is generally highest in the Group A picrites due to higher liquidus

temperatures (Verhoogen, 1962), and higher activity of Al_2O_3 in the liquid as a result of spinel and olivine fractionation only; group B dykes have, prior to final quenching, fractionated Al-bearing phases, namely spinel, plagioclase, clinopyroxene. Differences in cooling and crystallization rates (Walker *et al.*, 1976) between groups A and B may also be a relevant factor.

Within any one sample the groundmass pyroxenes have lower Al_2O_3-contents than the phenocrysts, in keeping with the zoning of the latter, and probably the result of decreasing pressure, temperature and activity of Al_2O_3.

Ti-Al relations (fig 4.15) reflect the spread in composition due to differences in cooling rate; the quenched samples are able to accommodate more Ti and Al (Shibata *et al.*, 1979). The Ti/Al ratios of 1/4 imply substitution of the type $R^{+2}(Fe,Mg,Mn) + 2Si^{+4} \rightleftarrows (Ti^{4+} + Fe^{+3} + Al^{+3}$, (Bence *et al.*, 1970; Grapes, 1975; Walker *et al.*, 1976), whilst the lower ratios, 1/6, are found in pyroxenes with higher Cr^{3+}-contents (Cr^{3+} competes successfully with Ti^{4+} for the octahedral sites; Nakamura and Coombs, 1973; Campbell and Borley, 1974).

Nickel in the pyroxene is just within (0.02-0.05 wt% NiO) the detection limit of the probe (see Appendix A) and minor amounts of Na_2O (0.64-0.30 wt%) are present throughout the fractionation range.

Pyroxene as an indicator of magma type: The chemical characteristics of the clinopyroxenes (high Ca, Ti, Al and limited Fe-enrichment) are similar to those typical of alkali basalts. However, in Si vs Al^{IV} (fig 4.16) and TiO_2 vs Al^{IV} (fig 4.17) plots, often used to discriminate between subalkaline (tholeiitic) and alkaline magma types, the Rhum data spread across all fields (even for a single sample); the majority fall in the transitional alkaline fields. In addition, the Ti- and Al-contents of clinopyroxenes have been shown to be unreliable indicators

of host magma composition. The effects of cooling rate (Walker *et al.*, 1976; Shibata *et al.*, 1979) are significant enough to confuse efforts to use pyroxene minor element chemistry to identify magma types: magma types can only be compared if they cooled at similar rates.

4.4.4 Opaque oxides

An extensive range of spinel solid solutions occurs in the Rhum dyke suite, as a result of the extensive substitution of Cr, Al, Fe^{3+} and Ti in the octahedral (B) sites, and Mg and Fe^{2+} in the tetrahedral (A) sites. The classification scheme used in this study is given in fig 4.18. The non-specific term "spinel" is used in its broadest sense to cover all compositions. Certain assumptions of stochiometry have been made in recalculating the analyses; (1) all Cr is assumed trivalent and in the B site with Al and Ti, (2) total iron is partitioned between A and B sites as Fe^{2+} and Fe^{3+}, in the proportions required for stochiometry (structural formula on the basis of 32 oxygens), (3) the possibility of site vacancies has not been considered. Analyses are presented in Appendix A, and the main compositional variation is between aluminous chromite and titanomagnetite.

Morphologically the spinels occur as euhedral or slightly rounded subhedral equant grains, typically 10-100 μm in diameter (maximum 500 μm) and varying in colour from pale brown to black. They have 3 modes of occurrence: (i) as homogeneous (unzoned) inclusions in olivine phenocrysts, (ii) as single, euhedral or slightly subhedral, microphenocrysts which are generally homogeneous (although zoning and sharply defined Fe + Ti-rich mantles are observed in some samples), and (iii) in the groundmass as anhedral magnetite grains and magnetite-ilmenite intergrowths. Types (i) and (ii) are absent from the Group B dykes.

4.4.4.1 Inclusions and phenocrysts: In the picrites (Group A), the majority of spinels (brown and black) occur as inclusions in olivine phenocrysts ($Fo_{92.5}$-Fo_{75}); some spinel microphenocrysts are also found. The spinels show a wide chemical variation, and significance of the data depends on whether the compositions are original (primary) liquidus compositions, or the result of secondary processes, e.g. reaction with liquid and/or subsolidus re-equilibration with adjacent silicate phases. In several samples the original chemistry of the spinels has been preserved by rapid quenching (B62/2, E53, M9). Spinels enclosed within olivine are assumed not to have undergone subsolidus Mg-Fe^{2+} cation exchange with the host olivine prior to quenching, as suggested by a lack of Mg/Fe zoning in both oxide and silicate phases. Spinel inclusions in those dykes which have not suffered rapid quenching, e.g. B89, M53, M21, B81, M190, may have undergone slight subsolidus re-equilibration, resulting in a slight decrease in Mg/Fe^{2+} ratio in the spinels, and an increase in Mg/Fe^{2+} in the host olivine (Irvine, 1967; Roeder *et al.*, 1979). However, in the majority of samples this effect is negligible since data lie on the trends defined by the quenched crystals; only two samples (M53 and M190) are sufficiently displaced from the general trends. Therefore, in conclusion, the spinel phenocrysts and inclusions are taken to represent a sequence of liquidus spinels whose chemical variation reflects the changes in bulk composition and intensive variables (temperature, pressure, f_{O_2}) of the magma during crystallization.

A positive correlation exists between the Mg/Mg+Fe composition of olivine and enclosed spinel (fig 4.19). Since this ratio decreases with decreasing MgO-content of the liquid (in the case of olivine), and decreasing temperature (for the spinel, Hill and Roeder, 1974; Fiske and Bence, 1980), the spinel Mg/Mg+Fe ratio can be used as an indicator of fractionation.

The compositional variation of the dyke spinels is shown in figs 4.20, 4.21 and 4.22; these represent projections onto the faces of a modified Johnstone spinel prism (Ti concentration is <1 cation per formula unit and is therefore ignored, Haggerty, 1976). There is an apparently unbroken range of compositions from Cr-pleonaste to Al-chromite to Al-Cr-magnetite to magnetite, for both phenocrysts and inclusions (fig 4.18 for nomenclature) and the complete range can be found within a single thin section.

The variation diagram of the trivalent cations Cr, Al, Fe (fig 4.20) reveals the presence of two trends, one showing variation in Cr/Al ratio ("Al trend"), and the other, marked variation in Fe^{3+}/Fe^{3+} + Cr + Al ratio ("Fe trend").

The Al-trend is shown by the translucent (brown) inclusion spinels (Al-Cr-picotites) in the magnesian olivines ($Fo_{92.5}$-Fo_{88}), and by microphenocrysts (Cr-pleonastes) in the quenched picrites; it involves a marked increase in Al^{3+} (with approximately constant Fe^{3+}) at the expense of Cr^{3+} (as shown by slight core to rim zoning). Henderson (1975) attributed a similar Al-trend (in the cumulus spinels of the ultrabasic complex) to postcumulus reaction of spinel with intercumulus liquid and cumulus silicates. This is clearly not the cause of the variation in the rapidly-quenched dykes; the Al-Cr-picotites and Cr-pleonastes are interpreted as high-pressure liquidus phases, having crystallized from primitive picritic (high-MgO) melts in equilibrium with mantle olivines ($Fo_{92.5}$-Fo_{89} - see section 4.4.1).

The Fe-trend is shown by opaque (black) spinel inclusions (in olivines Fo_{84}-Fo_{75}) and phenocrysts (Al-chromites and Al-Cr-magnetites) in quenched and non-quenched picrites, and involves marked enrichment in Fe^{3+} at the expense of Cr^{3+} and Al^{3+}. Curvature of the trend is due to decreasing Al/Cr ratio (Al^{3+} decreases more rapidly than Cr^{3+}),

and probably the result of one or more of the following: (i) Al^{3+} competing successfully with Cr^{3+} for the octahedral sites (unlikely in view of the high octahedral site preference energy of Cr^{3+}; Burns, 1970), (ii) faster rate of diffusion of Al^{3+} (relative to Cr^{3+}) towards the growing crystals, (iii) the earlier crystallization of Al-spinel depleting the liquid more rapidly in Al^{3+} than in Cr^{3+}; the most likely cause. The spinels of the Fe-trend are interpreted as low-pressure liquidus phases.

Fig 4.20 compares the dyke data with those from the layered ultrabasic complex (L.U.C.). Spinels included in L.U.C. olivines (Al-chromite and chromite) are characterized by significantly higher Cr^{3+}-contents, limited $Fe^{3+}+Al^{3+}$-enrichment, and show weakly-defined Al- and Fe-trends. L.U.C. spinels not enclosed by olivine show well-developed Fe- and "secondary Al-" trends, as a result of post-cumulus reaction with intercumulus liquid (trapped and non-trapped) and silicates (see Chapter 2). Primary (dyke) and secondary Al- trends are dissimilar, the latter being characterized by increasing Al^{3+} at constant or decreasing Cr^{3+}/Fe^{3+}.

A small compositional gap appears to separate the spinels of the dyke Al- and Fe- trends, and Cr-rich spinels typical of the L.U.C. are rarely found in the dykes.

Plots of Cr/Cr+Al against Mg/Mg+Fe are shown in fig 4.21. The aluminous spinels (Al-Cr-picotites and Cr-pleonastes) have a restricted range of Cr/Cr+Al ratios (0.58-0.40) and a wide range of Mg/Mg+Fe (0.70-0.34), and in this respect are more aluminous and magnesian than the lower pressure Al-chromites and Al-Cr-magnetites, (Cr/Cr+Al, 0.51-0.73 and Mg/Mg+Fe, 0.30-0.11). The two groups show different trends with fractionation; the aluminous spinels show decreasing Cr/Cr+Al, whilst the more iron-rich spinels show increasing Cr/Cr+Al. Zoning

of the phenocrysts is continuous and generally insignificant (magnitude rarely amounts to more than 0.05 units of Mg/Mg+Fe and Cr/Cr+Al), although in sample M9 some show a sharp mantle of Al-magnetite (maximum zonation 0.17 Mg/Mg+Fe and 0.11 Cr/Cr+Al). Inclusion spinels are unzoned.

The aluminous spinels from the quenched picrites plot within the fields of spinels from alpine peridotites, peridotite inclusions (after Irvine, 1967), oceanic mantle-derived peridotites (after Hamlyn and Bonatti, 1980) and quenched mid-ocean ridge (M.O.R.) basalts, fig 4.21, but are characterized by very restricted Cr/Cr+Al ratios and a greater range in Mg/Mg+Fe values (reflecting a range of conditions during crystallization and rapid cooling). For a given Cr/Cr+Al ratio they are less magnesian than spinels in M.O.R. basalts. They plot outside the fields of high-Cr podiform chromites, and overlap slightly with the more Al-rich compositions from stratiform intrusions. Inclusion spinels (Al-chromites) in the Rhum L.U.C. (Chapter 2) overlap with the Al-rich dyke spinels but range to lower Mg/Mg+Fe and higher Cr/Cr+Al ratios.

It has been suggested that Cr/Cr+Al ratios vary inversely with pressure (Bryan, 1972; Frey *et al.*, 1974; Dick, 1976; Sigurdsson and Schilling, 1976), but the occurrence of equally aluminous spinels in basalts believed to have crystallized at low-pressure (Table 4.3) suggests pressure is not the only factor involved. However, the failure by Fiske and Bence (1980) to synthesize high-Mg,Al spinels from experimental charges doped with Al, indicates that high activity of Al_2O_3 alone is not a major factor, and pressure must indeed be involved.

The Fe-trend spinels in the picrite dykes are (a) less magnesian (Mg/Mg+Fe 0.31-0.10), (b) generally have higher Cr/Cr+Al ratios (0.45-0.73, due to Al decreasing more rapidly than Cr) than the Al-trend

spinels, and (c) slightly overlap with the fields of spinels from stratiform intrusions (including the Rhum L.U.C.).

Variation in the Fe^{3+} cation-fraction for the dyke spinels is illustrated in fig 4.22. Both inclusions and phenocrysts define a continuous fractionation trend, with Fe^{3+}/R^{3+} ratios increasing slightly in the Al-rich spinels (0.06-0.14), then increasing sharply from the Al-chromites to Al-Cr-magnetites (0.22-0.94). Zoning of phenocrysts parallels the trend and is gradational and generally weak (av. 0.03 Fe^{3+}/R^{3+} increase); the exception is M9 (quenched picrite) where Cr-pleonaste crystals are sharply rimmed by Al-Cr-magnetite (maximum core to rim increase 0.42 Fe^{3+}/R^{3+}). Spinels from a variety of volcanic rocks have similar Fe^{3+}-contents, irrespective of host magma-type and depth of spinel crystallization, and similarly the Fe^{3+}-poor spinels plot in the fields of alpine peridotites, peridotite nodules and stratiform intrusions, irrespective of widely varying Cr/Cr+Al ratios (fig 4.22).

Discussion and conclusions: The spinels of the Rhum picritic dykes are unusual with respect to their apparent continuous gradation from Al-rich to Fe^{3+}-rich compositions, and it would therefore appear that spinel was a stable phase throughout the crystallization history of these rocks. Similar extensive chromite-titanomagnetite solid solution series have been recorded from Grenada alkali olivine basalts (Arculus, 1974) and the Elephant's Head Intrusion (Eales and Snowdon, 1979). Previous occurrences of spinel intermediate between chromite and magnetite have been ascribed to reaction of early-formed Cr-rich spinels with residual liquid during the end stages of crystallization (Gunn *et al.*, 1970; Evans and Moore, 1968; Evans and Wright, 1972; Ridley, 1974, 1977). However, the Rhum spinel phenocrysts and inclusions are taken to represent a sequence of liquidus spinels, whose

chemical variation reflects the changes in bulk composition and intensive variables (temperature, pressure and f_{O_2}) of the liquid during crystallization; those crystals included in olivine are assumed to have been prevented from reacting with the liquid and the majority of phenocrysts have had their original chemistry preserved by rapid quenching.

Controls over primary compositional variation in Cr-bearing spinels have been discussed by Hill and Roeder (1974), Ridley (1977) and Fiske and Bence (1980). Initial concentrations of Cr, Al, Fe^{3+}, Ti, Fe^{2+} and Mg reflect temperature and pressure in the zone of partial melting, and the mineralogy of the source material; subsequent variation in activities of these elements are a function of decreasing temperature, ?pressure, and structure of the melt.

Fig 4.23 illustrates that compositions of spinel in equilibrium with picritic/basaltic liquid change dramatically in response to decreasing temperature and Mg/Mg+Fe ratio in the liquid. Cr and Al decrease (the latter more rapidly) and Fe^{2+}, Fe^{3+}, Ti^{4+} and Fe^{3+}/Fe^{2+} (oxidation ratio) increase with decreasing temperature. The changes involve several cation exchanges: Fe^{2+} for Mg, Fe^{3+} for Cr and Al and a coupled substitution $Fe^{2+} + Ti^{4+}$ for Cr + Al.

Fe^{3+} increases with decreasing temperature and increasing f_{O_2} (Fiske and Bence, 1980) and because of the large octahedral site preference energy of Cr^{3+} (Burns, 1970) and none for Al^{3+} or Fe^{3+}, the increase in ferric iron reflects its substitution for Al^{3+} rather than Cr^{3+}. The Fe^{3+}/Fe^{2+} ratio remains constant and only decreases markedly once Al-Cr-magnetite crystallizes; it follows trends predicted by Hill and Roeder (1974) for crystallization under conditions of increasing f_{O_2}.

With respect to Cr-contents the liquidus spinels examined in this study are distinct from the more Cr-rich spinels found in the L.U.C.

(Chapter 2 and fig 4.20). Since the Cr-spinel stability field is very sensitive to small changes in the Cr-content of the liquid, and the solubility of Cr in basaltic liquid increases with increasing f_{O_2} and pressure (Hill and Roeder, 1974 - as a result of depolymerization of the silicate liquid creating octahedral sites), the limited crystallization of Cr-rich spinel in the dykes may be explained in terms of generation of a Cr-poor liquid at the site of partial melting - the low Cr-content being a function of the mineralogy of the source material, degree of partial melting, and conditions of low f_{O_2}. Alternatively the Cr-rich compositions of the L.U.C. spinels may be related to the appearance of plagioclase on the liquidus, since once plagioclase forms, the Cr-content of spinel increases markedly, due to the feldspar competing successfully for Al (Dickey and Yoder, 1972; Fiske and Bence, 1980); plagioclase was not a liquidus phase during crystallization of the picrites and therefore no marked increase in Cr is found. The overall decrease in spinel Cr^{3+}-contents with fractionation is due to decreasing Cr-content of the liquid (Akella *et al.*, 1976; Fiske and Bence, 1980) and increasing f_{O_2} (Hill and Roeder, 1974).

Al^{3+}-contents (fig 4.23) after an initial increase (for Al-trend spinels) show a marked decrease with falling temperature (for Al-poor spinels of the Fe-trend), i.e. Al/Cr increases, then decreases. The initial increase in Al/Cr is due to crystallization at temperatures above the liquidus for plagioclase (Dickey and Yoder, 1972) whilst the decrease resulted from the increasing substitution of Fe^{3+} (as f_{O_2} increased) and falling Al^{3+} concentration in the liquid.

Extreme compositional variation in sample M9 phenocrysts is almost certainly the result of a two-stage process; the high-Al spinel cores (crystallized at high pressure) are sharply mantled by Fe^{3+}-rich spinel identical in composition to the phenocrysts which have crystallized at

low pressure (Al-Cr-magnetite). The occurrence of spinels of intermediate composition, in picrite dykes of the same suite, precludes the possibility that the sharp contacts represent a compositional hiatus.

In conclusion, therefore, two chemically distinct groups of spinel phenocrysts and inclusions are present in the Rhum picrite dyke suite.

(i) the Al-rich spinels (Al-Cr-picotite and Cr-pleonaste) which show increasing Al/Cr (at constant Fe^{3+}) with decreasing temperature and define an Al-trend; they are believed to have acquired their high-Al_2O_3 (and MgO) characteristics during crystallization at high pressure, and been subsequently preserved by rapid ascent and quenching. The high-MgO content of the spinel-bearing picritic liquids, and the lack of significant zoning in the spinels suggests the magma did not suffer extensive fractionation during ascent from a high-pressure to a lower-pressure regime.

(ii) the Fe^{3+}-rich spinels (Al-Cr-magnetites and magnetites, together with the titanomagnetite in the groundmass - section 4.4.4.2) show increasing Fe^{3+} at the expense of Al and Cr, and are believed to have crystallized at much lower pressures and higher f_{O_2}.

The two groups of spinel represent a single series of liquidus compositions, in equilibrium with MgO-rich liquid, and whose chemistry changes dramatically in response to (a) decreasing pressure and temperature, (b) increasing f_{O_2}, together with (c) changing liquid composition (decreasing Mg, Al, Cr and increasing Fe^{2+}, Fe^{3+}, and Ti activities). Changes involve several cation exchanges, and generally suggest an extensive solid solution series exists between Cr-Al spinels and Fe^{3+}-Ti spinels.

The scarcity, within the picrite dykes, of Cr-rich spinels compositionally similar to those enclosed in olivine in the L.U.C. may imply

that the picritic liquids are not suitable parent liquids for the Rhum layered intrusion. However, the relatively Cr-poor nature of the dyke spinels may be accounted for by crystallization under conditions of higher pressure (Mg-Al couple is stable) and followed by higher f_{O_2}.

4.4.4.2 Olivine-spinel geothermometer: The olivine-spinel geothermometer was first treated theoretically by Irvine (1965, 1967). Jackson (1969) derived a geothermometer based on Mg-Fe^{2+} exchange equilibrium between the two phases in Stillwater cumulates, assuming ideal mixing; but when applied to olivine-Cr spinel pairs in volcanic rocks (Evans and Wright, 1972) the calculated temperatures were unrealistically high (ca. 2000°C). Evans and Frost (1975) suggested a more empirical approach to the problem, based on graphical calibration. In this study the methods of (a) Roeder *et al.* (1979) - involving a re-examination of Jackson's (1969) equation in the light of new thermodynamic data, and of (b) Fabries (1979) (a calibration of the Evans and Frost (1975) geothermometer), are used.

Use of olivine-spinel equilibrium as a geothermometer requires knowledge of (a) the absolute free-energy of formation of the end-member spinels and olivines for the operative temperature range, and of (b) the mixing behaviour of olivine and spinel solid solutions over that temperature range. With regard to (b), mixing within the A and B structural sites of spinel is assumed ideal, although interactions between the two sites are unaccounted for (Wood and Nicholls, 1978). Data on (a) are not known in sufficient detail; a range of free-energy values is reported in the literature and it is apparent that this has a significant effect on the calculated equilibration temperatures (differences of up to 1000°C are reported by Roeder *et al.*, 1979).

The majority of theoretical and experimental work has concentrated on Cr-rich spinels, for which the free-energy values are best known.

However, in the dykes the inclusion spinels have Fe^{3+}- and Al^{3+}-rich chemistries, for which the free-energy values are not well established.

Calculated equilibration temperatures for a number of olivine-spinel pairs in the picritic dykes (Group A) are listed in Table 4.4. Differences of up to 200°C are found between the two methods used, and the magnitude of uncertainty that can be introduced in the temperatures due to analytical errors in the determination of Fe and Mg in olivine and Cr, Al and Fe^{3+} in spinel is approximately ±50°C (Jackson, 1969). Temperatures calculated for the Al-rich spinels (M9, B62/2, E71) range from 487-752°C and 699-887°C (using Roeder *et al.*, 1979 and Fabries, 1979, respectively), and in the rapidly-quenched samples (M9 and B62/2) tend to show a large and unreasonable range of values, far below the expected near-liquidus temperatures. Subsolidus re-equilibration of Mg and Fe cannot account for the discrepancy (it is not expected in quenched rocks) and it is concluded that the Al-rich spinels may only be useful as a geothermometer when the free-energy values for the Al end-member spinels are better known. However, the possibility must also be considered that the mineral pairs may be actually unequilibrated.

The Fe^{3+}-rich spinels yield calculated temperatures of 332-470°C (Roeder *et al.*, 1979) and 419-596°C (Fabries, 1979). These spinels occur in the relatively coarse-grained, non-quenched samples (M190, M21, B89), which therefore in all probability have suffered extensive Mg-Fe^{2+} re-equilibration to temperatures well below the solidus (Roeder *et al.*, 1979) as a direct result of their intrusion into a layered complex which probably took thousands of years to cool. However, Roeder *et al.* (1979) concluded that Fe^{3+}-rich spinel is relatively insensitive to temperature, and is not useful as a geothermometer since accurate analyses are required in order to obtain accurate

Fe^{3+}:Fe^{2+} estimates from stochiometry, and to decrease the sensitivity of $K_{D_{Fe-Mg}}^{sp-ol}$ to errors in Fe^{3+}.

In conclusion, the Al^{3+}- and Fe^{3+}-rich compositions of the dyke spinels are not conducive to use as geothermometers. Until more accurate free-energy values for these Al and Fe^{3+} end-members are known, the absolute calculated temperatures of re-equilibration are suspect.

4.4.4.3 Groundmass opaque oxides: Groundmass Fe-Ti oxides are ubiquitous throughout the Rhum dyke suite; they comprise interstitial titanomagnetite grains with trellis, sandwich and composite ilmenite intergrowths, together with homogeneous ilmenite grains in contact with, or independent of, titanomagnetite. In all dykes they have clearly crystallized at a late stage. Owing to the generally very fine grain-size of the oxides, limited chemical data have been collected. Analyses are listed in Appendix A.

Ilmenite: Ilmenite which crystallized originally as discrete grains is termed "primary"; the lamellae and laths of trellis, sandwich and composite ilmenite are exsolved generations. Analyses are plotted in fig 4.24. Iron-contents have been calculated into wt% FeO and Fe_2O_3, and compositions calculated in terms of mol % end-members (both according to Carmichael, 1967). Compositions range from $Il_{99.2}\ Hm_{0.8}$ to $Il_{89}\ Hm_{11}$ and are comparable to those from other basic intrusives and extrusives (Haggerty, 1976). Although many of the grains, both primary and exsolved, appear homogeneous and unoxidized, the majority have suffered slight oxidation, and therefore plot to the TiO_2 side of the Il-Hm join (fig 4.24).

Several minor elements (Cr, Ni, Al, Mg and Mn) were also analysed. Contents of Cr_2O_3, NiO and Al_2O_3 are low and range 0.0-0.81 wt%, 0.0-0.15 wt% and 0.0-1.35 wt% respectively. MgO (geikilite component) varies from 0.27-8.77 wt% and contents are similar to those of ilmenites

in the Rhum gabbros (0.14-5.75 wt%), allivalites (0.5-10.4 wt%) and peridotites (1.8-9.2 wt%) - see fig 3.27. The abundance of MgO in the parental liquid, and the amount of olivine fractionation are constraints on Mg-entry into ilmenite (Haggerty, 1976). Contributory factors include the rate and path of equilibration, since in lunar rocks ilmenite is known to equilibrate with coexisting silicates (and therefore change composition with decreasing temperature, Usselman, 1975), and since Mg preferentially partitions into ilmenite rather than coexisting magnetite (Stormer, 1972).

MnO (0.24-4.65 wt%, fig 4.24) is strongly partitioned towards ilmenite in ilmenite-magnetite intergrowths, regardless of whether it is primary or exsolved (Czamanske and Mihalik, 1972; Neumann, 1974).

Since, on textural grounds, it is often difficult to distinguish between primary and secondary ilmenite generations, mineral chemistry is a useful indicator of type (as suggested by Evans and Moore, 1968; Lipman, 1971; Duchesne, 1972 and Mathieson, 1975). Differences between the primary and exsolved dyke ilmenites are similar to those reported from the gabbros (section 3.4.4.1).

Titanomagnetite: Titanomagnetite analyses are listed in Appendix A and plotted in fig 4.24. Fe_2O_3-contents are calculated according to Carmichael (1967) and mol % Usp and Mt by the method of Powell and Powell (1977).

Compositions range from $Usp_{7.8}\ Mt_{92.2}$ to $Usp_{38.2}\ Mt_{61.8}$ and extend to slightly more Usp-rich compositions than grains from the gabbros (Usp_4-Usp_{23}). The majority of grains have been oxidized and therefore do not represent primary high-temperature magmatic compositions. The process of oxidation induces a characteristic partitioning of the minor elements; MgO (0.11-2.56 wt%) enters the ilmenite, Al_2O_3 (0.32-9.72 wt%) and Cr_2O_3 (0.09-12.76 wt%) prefer the spinel structure. MnO-contents

(0.13-0.67 wt%) are usually higher in the titanomagnetite (fig 4.25). Variation in TiO_2 (2.79-13.67 wt%) is a function of TiO_2-levels in the primary spinel, and the degree of "contamination" of the microprobe beam spot by fine trellis ilmenite lamellae, whilst CaO and SiO_2 are attributed to silicate contamination.

In fig 4.26 analyses are plotted in a ternary projection of the multicomponent spinel prism, and it is evident that compositions are a continuation of the Fe-trend displayed by the phenocrysts and inclusions. Hill and Roeder (1974) have shown that spinel crystallization is continuous from chromite to titanomagnetite for f_{O_2}'s $>10^{-8.5}$ atm, and that Mg-, Al- and Cr-bearing titanomagnetites are stable under conditions of low temperature and constant f_{O_2}, or increasing f_{O_2} at constant temperature.

Ilmenite-titanomagnetite geothermometry and oxygen barometry:

The equilibrium temperature and f_{O_2} of coexisting magnetite and ilmenite can be determined from their compositions (Buddington and Lindsley, 1964). Compositions of the fast-quenched Fe-Ti oxides (samples M9, B62/2, E53) which would be expected to give liquidus temperature and f_{O_2} (Carmichael, 1967) could not be determined due to their very fine grain-size; data from the more slowly cooled dykes (B89, B65, B2) yield lower temperatures and f_{O_2}'s because of subsolidus re-equilibration (Anderson, 1968; Duchesne, 1972; Bowles, 1976, 1977; Rollinson, 1979).

Analyses from ilmenite-titanomagnetite intergrowths in three dykes are presented in Table 4.5. Of the various methods used to reallocate total iron into FeO and Fe_2O_3, and minimize the effects of minor RO and R_2O_3 components (reviewed by Bowles, 1977), that of Carmichael (1967) was used. Estimates of Usp-fractions were obtained using the relation wt% $TiO_2/35.75$ (Powell and Powell, 1977). Temperatures and f_{O_2}'s obtained from the calibration curves of Buddington

and Lindsley (1964) for the recalculated present compositions of titanomagnetite and ilmenite are listed in Table 4.5 and plotted in fig 4.27. The temperatures (610-730°C) and f_{O_2}'s ($10^{-16.3}$-$10^{-20.3}$atm) define a curve parallel to, and coincident with, the QFM buffer curve, and their ranges are higher than those of the gabbro Fe-Ti oxides (540-600°C and $10^{-23.5}$-$10^{-19.3}$ atm). Temperatures represent "blocking-temperatures" (i.e. the temperature below which diffusion was unable to form the exsolved ilmenite) and are a function of the rate of diffusion and amount of exsolved phase present (Rollinson, 1979). The trellis-ilmenite generation is too fine to analyse and therefore final equilibration conditions could not be determined.

Using the method of Mathieson (1975 - outlined in section 3.4.4.3), the compositions of the original high-temperature Fe-Ti oxides can be estimated, and hence the variation of temperature and f_{O_2} during crystallization. The inferred values are listed in Table 4.6 and plotted in fig 4.27; the original compositions plot close to the QFM buffer curve in the range 920-1020°C and f_{O_2} 10^{-11}-$10^{-13.5}$ atm (these figures are subject to experimental errors of ±30°C and ±10^{-1} atm in the calibration curves, Buddington and Lindsley, 1964). Values are higher than those given by the gabbros (875-900°C and 10^{-12}-$10^{-13.5}$ atm) - fig. 4.27.

4.4.5 Amphibole

Analyses of primary amphiboles from the Rhum dykes are listed in Appendix A. In the absence of Fe^{3+}, OH and F, the analyses have been recalculated on the basis of 23 oxygens, all Fe treated as Fe^{2+} and site occupancies calculated according to Leake (1978). The majority of amphiboles are sufficiently low in Fe^{2+}/Fe^{2+}+Mg and Fe^{3+}/Al^{VI} (at fixed oxidation ratio) and high in Ti (>0.5 atoms) to be kaersutites. Compositions with <0.5 Ti atoms are pargasites.

Although $Mg/Mg+Fe^{2+}$ ratios (0.81-0.74) indicate a limited degree of fractionation, the minor elements show more pronounced variations. In the kaersutites TiO_2 and Al^{IV} increase with iron-enrichment (5.16-6.51 wt% and 1.74-1.97 cations respectively) and K_2O decreases (0.74-0.55 wt%). Increasing TiO_2 suggests the kaersutite crystallized prior to the Fe-Ti oxides. In contrast, fractionation trends for the pargasites ($Mg/Mg+Fe^{2+}$ 0.81-0.77) involve slightly decreasing TiO_2 (0.48-0.46 wt%) and decreasing Al^{IV} (1.83-1.70 cations). No zoned kaersutite or pargasite was found.

These silica-undersaturated amphiboles are typically associated with basic undersaturated rocks (alkali olivine basalts and related rock types) e.g. Vincent (1953), Aoki (1963, 1970), Brown (1973), Lewis (1973), Brooks and Platt (1975), Grapes (1975), Baxter (1978), although Helz (1973) suggested this may be a consequence of their higher volatile fugacity, relative to tholeiites, rather than any direct result of the undersaturation. Alternatively, Yagi *et al.* (1975) attributed some kaersutite rims and replacements of titanaugite to a reaction relation between pyroxene and liquid at temperatures between 950°C-1050°C. Generally TiO_2 in the adjacent pyroxenes is too low (<1 wt%) for this to apply, and the Rhum dyke kaersutites therefore probably crystallized directly from Ti-enriched, silica-undersaturated interstitial liquid; their formation instead of pyroxene was due to an increase in the volatile-content of the liquid.

Compared to kaersutites with similar TiO_2-contents (Brooks and Platt, 1975; Grapes, 1975; Donaldson, 1977b) the Rhum grains have higher $Mg/Mg+Fe^{2+}$ ratios, higher Cr_2O_3, and lower K_2O, Al^{IV}, and Al^{VI}.

4.4.6 Mica

Microprobe data for the micas are given in Appendix A. All Fe was treated as FeO, analyses recalculated on the basis of 23 oxygens,

and cations assigned as suggested by Deer, Howie and Zussman (1963). Tetrahedral (Al + Si) totals range 7.52-8.25 cations and only three analyses show Si + Al deficiencies: similar deficiences were reported by Nash and Wilkinson (1970) for the Shonkin Sag intrusion. With Mg: Fe ratios >2:1 and high TiO_2-contents (4.26-7.86 wt%) the micas are "titanphlogopites"; identical compositions are reported from the Blue Mountains alkaline ultrabasic complex (Grapes, 1975).

Atomic proportions of the most significantly variable elements are plotted on fig 4.28. The phlogopites have a limited range of Mg/Fe + Mg ratios and Fe/Fe + Mg with fractionation (directions of biotite evolution in the Klokken intrusion (Parsons, 1979) are shown for comparison). MnO shows a steady increase (0.03-0.09 wt%), and Cr_2O_3 a decrease (0.29 – 0 wt%), with increasing Fe-content. The low Cr_2O_3-contents suggest the phlogopites crystallized from late interstitial liquid, and were not the products of reaction between liquid and Cr-bearing clinopyroxene (±Cr spinel). The high TiO_2-contents reflect the TiO_2-rich nature of the late-stage interstitial liquid, and are not believed to be the result of reaction between liquid and the Fe-Ti oxides (which they often rim). In summary, the phlogopites are the direct crystallization product of undersaturated TiO_2-rich residual liquid (in many cases together with kaersutite).

4.5 Summary

1. Dykes within the L.U.C. are generally <1 m wide.
2. Two main types are present: picrites (rich in olivine phenocrysts, up to 60%; and often with quenched groundmasses) and more abundant basalts (extremely fine-grained; containing sparse microphenocrysts of olivine, plagioclase and clinopyroxene).

3. Rare syenitic aplites are recorded.

4. Olivine phenocrysts range $Fo_{92.5}$-$Fo_{72.5}$ (those >Fo_{89} are of high-pressure origin) and groundmass grains Fo_{84}-Fo_{70}: all are Ni-depleted due to sulphide fractionation.

5. Plagioclase phenocrysts vary from An_{89}-An_{55}; groundmass laths An_{76}-An_{43}.

6. Clinopyroxene phenocrysts (Cr-rich diopside and calcic augite) have crystallized at low pressure; groundmass grains are Ti-rich and Al-rich diopside, salite and calcic augite.

7. Phenocryst and inclusion (within-olivine) spinels show a wide compositional range: two distinct groups are present :- Al-rich picotite and pleonaste (high-pressure liquidus phases), and Fe^{3+}- and Cr-rich low-pressure liquidus phases (Al-chromite and Al-Cr-magnetite).

8. Groundmass Fe-Ti oxides (titanomagnetite and ilmenite) yield late-stage magmatic temperatures and f_{O_2}'s of 920-1020°C and 10^{-11}-$10^{-13.5}$ atm respectively.

9. Primary hydrous phases include kaersutite and Ti-rich phlogopite.

CHAPTER 5

THE PERIDOTITIC MINOR INTRUSIONS

5.1 Introduction

Several intrusive bodies of peridotite, ranging from dunite to feldspathic peridotite, are found within the layered ultrabasic complex (ultramafic nomenclature as in Chapter 2).

To date, peridotitic minor intrusions have only been described from outside the ultrabasic complex (Harker, 1908; Dunham, 1965 ; Dunham and Emeleus, 1967; Emeleus and Forster, 1979). For example numerous elongate masses (up to 700 m long) occurring in the Torridonian arkoses to the north of the Main Ring Fault (fig 1.4) have been regarded as defining a large ring structure (Bailey, 1945), and as part of a radial system centred on Glen Harris (Dunham and Emeleus, 1967). Most plugs have intensely metamorphosed the adjacent arkoses: this, together with the occurrence of basaltic "envelopes" around some plugs (Dunham and Emeleus, 1967) may indicate that the original pipes were initially occupied by nearly-aphyric basalt, before being filled by a highly-porphyritic magma or crystal mush. One small plug (NG 35550092) contains abundant small (0.2-0.5 m) subrounded dunite fragments (Emeleus and Forster, 1979).

Along the northern margin of the ultrabasic complex, to the east of the Long Loch Fault, Dunham (1965) mapped three dyke-like masses or "tongues" of feldspathic peridotite extending northwards from the layered series (fig 1.4). They cut Lewisian gneiss, Torridonian arkose, explosion breccia, granophyre and the Main Ring Fault (eastern tongue only), and are distinguished from the L.U.C. peridotites by the vertical banding developed in the interstitial material (termed "matrix-banding", Dunham, 1965). The western extension of the Inner Ring Fault separates the tongues from the layered series: the tongues

are therefore earlier than emplacement of the L.U.C.

The apparent lack of similar peridotite masses in the Eastern and Western Layered Series may reflect the difficulty of locating plugs in petrographically similar host rocks.

5.2 Field Relations: This Study

The Eastern Layered and Central Series ultrabasic rocks in the Barkeval-An Dornabac area are invaded by a number of peridotitic minor intrusions (Map 1). A detailed study has been undertaken with a view to (a) describing their mineralogical and chemical variation, (b) establishing chronological relationships, and (c) ascertaining the genetic relationship, if any, between them and the main layered complex. Peridotitic masses from the Salisbury's Dam area, and the "tongues" have been re-investigated, and peridotite from the Sgaorishal and Kinloch Glen plugs included for comparison.

5.2.1 The dunite plugs

Three elongate dunite intrusions occur within the westernmost exposures of the E.L.S.: 150 m SE of Triangular Loch (378977); 400 m SE of Priomh Lochs (372983); and 700 m SE of Long Loch dam (367977). Generally elliptical in outcrop, they vary in size from 500 x 100 m to 100 x 40 m, and form a series of low rounded knolls whose pale buff-coloured, smooth weathered surfaces contrast with the dark brown, heavily-pitted layered peridotites. Intrusive contacts are approximately vertical, and the dunite neither veins nor brecciates the host ultramafites.

The dunites are homogeneous and fine-grained, but contain concentrations of interstitial feldspar and clinopyroxene producing ill-defined patches of feldspathic and pyroxene-rich peridotite. Internal structures are rare: igneous layering is absent, the only fabric

(sporadically developed in the central portion) being defined by concentrations of Cr-spinel in the form of wispy, discontinuous 2 mm thick "layers". Near the margins, a distinctive weathering phenomenon, in the form of narrow, sinuous, fairly continuous vertical ridges (amplitude 0.5 cm and 2-3 cm apart), is the result of preferred orientation of tabular olivines. In general the dunites are weakly to moderately serpentinized (<30% serpentine products).

5.2.2 The peridotite tongues

The three masses of peridotite extending northwards from the L.U.C. (east of the Long Loch Fault) are, from west to east, the Long Loch, Priomh Lochs and Loch Duncan tongues (Map 1), and measure ca. 700 x 360 m, 450 x 250 m and 600 x 150 m respectively. The two western tongues form extensive areas of moderate to low relief, comprising small, isolated knolls of dark greenish-brown peridotite, whilst in contrast, the much higher relief Loch Duncan mass forms a series of large rounded, dun-coloured exposures with a very characteristic "tortoise-shell" jointing pattern. Mapping confirms lithological and structural (internal) differences between the masses.

5.2.2.1 The Long Loch tongue: Varying proportions of interstitial feldspar and clinopyroxene result in a wide range of weathering characteristics, from smooth dunite to coarsely-pitted pyroxene-rich peridotite: the rocks are moderately serpentinized. Grey-black olivines are subhedral to euhedral (1-2 mm), and lack a preferred orientation. Occasional irregular patches contain coarser grains (up to 5 mm): some show a distinct bimodal size distribution. No internal contacts have been found.

The variation in relative proportions of interstitial feldspar and pyroxene, together with slight changes in olivine grain size, define internal layering. Three main types have been found:

(a) irregular lenses and wedges (av. 0.1 x 1 m) of fine-grained dunite, rich in disseminated Cr-spinel, enclosed within coarser-grained feldspathic peridotite, (b) interstitial feldspar locally concentrated into discontinuous "layers" (1-2 cm thick) and (c) 0.2-2 m thick layers of coarsely-pitted feldspathic peridotite, alternating with less-pitted, more wispily-banded dunitic peridotite.

All three types dip southwards, shallowing progressively from 56° at the northern end to 8° at the south. No vertical layering has been found parallel to the margins.

5.2.2.2 The Priomh Lochs tongue: This, the smallest of the three, is continuous with the Long Loch tongue. It also comprises a moderately serpentinized peridotite, rich in blackish olivines, and with variable proportions of interstitial silicates. Clinopyroxene, the dominant interstitial phase, imparts a characteristic knobbly and pitted surface texture. Internal structures are rare; only a weak type (c) layering is sporadically developed, which defines a shallow N-S trending syncline in the central region of the tongue.

5.2.2.3 The Loch Duncan tongue: The southernmost 200 m comprises peridotite similar to that of the adjacent tongues: the remainder forms a separate dyke-like intrusion, which is distinctly more olivine-rich, and finer-grained, and displays distinctive "matrix-banding".

As described by Dunham (1965) this banding (defined by alternating light and dark peridotite) is generally horizontal on the uppermost exposures, and steeply inclined parallel to the margins over much of its length. However, on closer examination, a more complex geometry is evident. The style of layering varies greatly (figs 5.1-5.4) and several additional sets of layering are present: (iii) one strongly-developed set dipping moderately (28-32°) to the east, (iv) a weaker set dipping gently (10-20°) to the west, and (v) a weak

set dipping steeply (64-70°) to the west. Sets (iii) and (iv) often intersect (fig 5.4).

The five sets cannot be adequately explained by Dunham's (1965) model of periodic undercooling of a eutectic pore fluid. Since they are better correlated with concentrations of chlorite and sericite, than with changes in relative proportions of interstitial feldspar and pyroxene, it is suggested that they are a form of jointing. Circulating hydrothermal fluids, rather than filtering uniformly through the pile, migrated into restricted irregular channels (partly controlled and defined by preferred orientation of the olivines).

5.2.3 The black peridotites

In the summit area and south face of Barkeval, several small elliptical peridotite plugs (200 x 80 m to 15 x 10 m) are easily distinguished from the E.L.S. and C.S. host peridotites, by their distinctive black olivines. Exposed contacts are vertical. In places the black peridotite has enclosed several small (7-17 cm) rounded fragments of adjacent gabbro, allivalite and olivine cumulate. Internal structures are absent.

5.3 Petrography

5.3.1 The dunite plugs

The olivines are generally equidimensional, 0.5-1 mm, subhedral to anhedral, and form a mosaic of interlocking grains whose straight or curved mutual boundaries meet in triple junctions. Occasional larger grains (up to 1.5 mm) display undulatory extinction and, or, deformation lamellae.

Clinopyroxene is a minor intercumulus phase (ca. 2%) forming irregular wedge-like grains between olivines, or larger (1.5 mm) equant grains poikilitically enclosing small (0.2 mm) rounded olivine

crystals. Intercumulus plagioclase is rare (<1%): when present, several zoned platelets occupy a single interstitial space. Red-brown amphibole (1-2%) poikilitically encloses very small olivines, rims disseminated spinels, and appears to have partially replaced clinopyroxene. Up to 8% (av. 4%) disseminated spinel is present, and the euhedral to subhedral grains (0.05-0.5 mm) form clusters between olivine, and are rarely enclosed in olivine. Occasional larger grains contain inclusions of red-brown amphibole and biotite.

5.3.2 The peridotite tongues

Petrographically the Long Loch and Priomh Lochs tongue peridotites (olivine-rich meso- and heteradcumulates) are similar to those of the Long Loch and Ruinsival Members of the Central Series. Relative to the E.L.S. cumulates they show greater abundance of (i) olivine, (ii) serpentinization, (iii) biotite and amphibole, (iv) late-stage chlorite and ?zeolites, together with a more restricted range of olivine morphologies.

Olivines show a wide range in grain-size (0.5-4 mm, av. 1.5 mm) and vary from predominantly subhedral/anhedral (Priomh Lochs tongue, e.g. J13) to euhedral (Long Loch tongue, e.g. J10): skeletal and tabular morphologies are rare. Several samples, e.g. B10, contain two olivine populations, (a) large serpentinized subhedral (5-8 mm), and (b) smaller (<1 mm) more abundant, euhedral-subhedral, less-serpentinized grains.

Due to varying degrees of postcumulus overgrowth, olivines show both grain-supported (Priomh Lochs tongue) and non grain-supported fabrics (Long Loch tongue). Large (4-10 mm) pale green clinopyroxene plates are poikilitic towards olivine, and occasionally subophitic towards feldspar. Subordinant, often markedly zoned, intercumulus plagioclase has a non-poikilitic texture: several small crystals occupy each pore space.

Crystallization products from late-stage (H_2O-rich) residual liquids include red-brown amphibole and biotite (the latter occasionally enclosing fine apatite-needles); they rim olivine, spinel, and clinopyroxene. The mafic phases have suffered variable marginal hydrothermal alteration to chlorite-magnetite intergrowths.

Olivines in the finer-grained (0.2-0.6 mm) more dunitic Loch Duncan tongue are often tabular, with a platy lamination parallel to the "matrix-banding". Larger grains (8 x 0.2 mm) are often bent and broken. Chloritization is patchy: maximum concentrations appear to correspond to the dark brown matrix-bands. The dark and light coloured bands cannot be directly correlated with dominance of either interstitial pyroxene and feldspar (as suggested by Dunham, 1965b), and an alternative origin has been suggested in section 5.2.2.3.

5.3.3 The black peridotite plugs

Of all the peridotitic minor intrusions, these most closely resemble the E.L.S. olivine cumulates. Closely-packed subhedral, equant olivines (2-3 mm) are set in small (<1 mm) plates of zoned plagioclase (sometimes cloudy), and subordinant green or brown clinopyroxene. As a result of high-temperature oxidation, the olivines are heavily clouded with two sets of micron-sized oriented dendritic platelets of a black opaque phase ?iron oxide; a similar, but much less concentrated, oxidation texture is common in E.L.S. olivines (Chapter 2). The majority of olivine is undeformed: deformation lamellae are rare.

Minor amounts (2.5%) of disseminated spinel are present as (a) rare subhedral rounded (0.2 mm) inclusions in olivine, surrounded by a narrow zone free of dendritic platelets, (b) coarse (0.4 mm) euhedral-subhedral grains between olivines, and (c) "trains" of very fine (<0.1 mm) euhedral-subhedral grains lying consistently along the same side of the olivines. Type (c) may be interpreted as a "geopetal"

structure, implying that some settling of suspended crystals took place during or after emplacement. The peridotites are fresh: olivine shows little serpentinization and chloritization, sericitization of feldspar is rare, and the pyroxene is occasionally incipiently altered to red-brown amphibole.

5.4 Mineral Chemistry

Electron microprobe analyses of the major silicate and oxide minerals are presented in Appendix A.

5.4.1 Olivine

Olivines in the tongues and dunite plugs show similar compositional ranges ($Fo_{85.6}$-Fo_{91}); more iron-rich compositions are found in the black peridotite and Salisbury's Dam masses (Fo_{83} and Fo_{80-81} respectively). All olivine is unzoned and the maximum within-sample variation is 1.5 mol% Fo. A comparable compositional variation is shown by the L.U.C. (Fo_{79}-$Fo_{89.4}$).

CaO-contents (0.02-0.38 wt%) show no correlation with Fo-content (fig 5.5). Within-sample variations are large (maximum 0.24%). Phenocrysts in the picritic dykes have comparable contents (0.02-0.44%, fig 4.4); cumulus olivines in the L.U.C. are considerably poorer in CaO (<0.12%, fig 5.5). Initially high-CaO olivines may have been reduced to lower values by (a) diffusion of Ca from olivine to clinopyroxene and plagioclase during slow cooling (Ferguson, 1978), and (b) depletion during serpentinization (Coleman and Keith, 1971).

MnO-contents (0.14-0.31%) show a steady linear increase with iron-enrichment (fig 5.6), and overlap considerably with the ranges given by dyke phenocrysts and L.U.C. cumulus grains.

Fig 5.7 illustrates the wide range in NiO-contents (0.18-0.42%) and the strong positive correlation with Fo. The data can be interpreted

in terms of fractionation from a sulphide-saturated magma (see Chapter 2): the most Ni-depleted olivines are found in sulphide (pentlandite)-bearing samples (M42 and M50). Similar NiO levels are shown by picritic dyke phenocrysts (0.49-0.2%); olivines from the L.U.C. are more markedly Ni-depleted.

5.4.2 Pyroxene

Clinopyroxene falls within the diopside field of the pyroxene quadrilateral and shows a more limited compositional range (Ca_{48} $Mg_{46}Fe_6$ - Ca_{46} Mg_{44} Fe_{10}) than intercumulus pyroxene from the L.U.C. (fig 5.8).

Al, Ti, Cr and Mn show considerable variation and a correlation with major element abundances (figs 5.9 and 5.10): the diopsides are rich in Cr (Cr_2O_3-contents, 1.5-0.7 wt%, are similar to those of the L.U.C. intercumulus pyroxene, 0.5-1.35%), Al (2.6-5.5 wt% Al_2O_3) and Ti (0.44-2.26 wt% TiO_2). The limited magmatic differentiation involved impoverishment of the liquid in Cr, and slight enrichment in Mn, Ti and Al.

Large within-sample minor element variations (maximum 0.5% Cr_2O_3; 1.7% Al_2O_3; 1.1% TiO_2) suggest partial or complete entrapment of liquid by the olivines, and poor diffusional contact between liquid in the different pores crystallizing clinopyroxene. The effects of subsolidus re-equilibration between the pyroxene and intercumulus plagioclase and cumulus Cr-, Al-, Ti-rich spinel (Chapter 2) may be superimposed on the compositional variation outlined above.

5.4.3 Feldspar

The intercumulus plagioclase shows a restricted compositional variation ($An_{82.6}$-An_{52}, majority An_{79}-An_{66}) and is generally less calcic than the poikilitic feldspar of the L.U.C. (An_{92}-An_{74}). Marginal zoning is limited (av. 3 mol% An, maximum 11%) compared to the often >10% in the latter.

Minor element abundances (FeO, 0.19-0.74%; MgO, 0-0.31 wt%; TiO_2, 0.03-0.18 wt%) are similar to those reported from the L.U.C., gabbros and dykes (Chapters 2, 3 and 4).

5.4.4 Spinel

Cr-spinel is disseminated throughout the peridotitic minor intrusions in an amount estimated at 1-3% (maximum of 10% locally). Textural features such as mutual interference and sintered appearance of contiguous grains are uncommon; the majority of grains are either subhedral and show no sign of postcumulus reaction, or are rounded and embayed, suggesting resorption has occurred prior to, or during, the postcumulus stage. All textural varieties may be observed in one thin-section (even side by side), and indicate the diverse influence the locally surrounding liquid and crystallizing phases may have on the spinel (see Chapter 2).

The compositional variation of Cr-spinel (illustrated in figs 5.13 and 5.14) is limited. Those within olivine (inclusion type) are assumed basically unchanged since time of inclusion (see Chapter 2). In terms of trivalent-cation substitution (fig 5.13) the inclusion spinels show similar Cr/Al variation to the equivalent grains from the layered series peridotites; however, grains within intercumulus silicates do not show the extensive Cr/Al and Fe^{3+}/R^{3+} variations typical of the layered cumulates (the secondary Al- and the Fe^{3+}-trends respectively, Chapter 2).

Variation in the Mg-cation fraction (Mg/Mg + Fe), fig 5.14, is also small (0.23-0.43 for inclusion spinels and 0.28-0.58 for disseminated grains). Cr/Cr + Al (0.46-0.83) and Fe^{3+}/R^{3+} (0.09-0.42) ratios for both spinel types fall within the fields for stratiform intrusions (fig 5.14).

Despite the absence of compositional zoning, neighbouring grains may differ substantially in composition, a result of interaction with surrounding liquid and, or, crystallizing phases (Cameron, 1975; Henderson, 1975; this study Chapter 2). However, the restricted spinel variation (relative to those in the L.U.C.) is regarded as a function of the smaller volumes of interstitial liquid allowing only limited opportunities for postcumulus reaction.

5.4.5 Amphibole and mica

The primary hydrous phases are Ti-rich phlogopitic mica (6.0-6.75 wt% TiO_2) and titaniferous calcic amphibole (pargasite and kaersutite, Leake, 1978) with 4.24-6.37% TiO_2 (see Appendix A). Both have crystallized directly from Ti-enriched, silica-undersaturated residual liquid.

5.5 Discussion

The peridotitic minor intrusions are modally (Table 5.1), texturally (section 5.3) and mineralogically (section 5.4) similar to the unlayered peridotites of the Central Series (Outer Breccia Zone and Ruinsival Members): they are significantly different in modes and textures from the peridotites of the E.L.S. (section 5.3). All occur within 300 m of the Central Series peridotites (detailed mapping may reveal similar masses within the adjacent Ard Mheall Member), and it is tentatively concluded that they are intrusive offshoots from the Central Series olivine cumulates.

Their lack of deformational features (e.g. intense marginal shearing) precludes emplacement in a solid state. Absence of chilled margins and evidence for *in situ* crystallization and differentiation (e.g. comb-layered harrisite, and fine-scale layering) suggests the masses formed by the intrusion of a highly porphyritic magma

("crystal-mush"), carrying at least 60% olivine (+spinel) crystals (see Chapter 2). The irregular lensoid concentrations of Cr-spinel may have resulted from flow segregation due to differential movement within the deforming crystal-liquid mush: presence of only slight olivine deformation suggests emplacement was slow. The relative abundance of primary hydrous phases (compared to E.L.S. olivine cumulates) is related to the nature of the magma: volatiles would be more efficiently trapped in a crystal-mush.

Emplacement of the crystal-mushes may have been analagous to emplacement of clastic intrusions associated with contemporaneous sedimentation, faulting and basinal subsidence (Eisbacher, 1970). Discrete displacements within the chamber floor, propagated upwards by means of contemporaneous faults within unconsolidated or semi-consolidated Central Series cumulates, initiated mobilization of crystal-mush. Mobilization and intrusion may have been forcibly upwards, or involved downward movement by simple filling of tension fissures ("neptunean dykes").

The lack of cross-cutting relationships between the various peridotitic minor intrusions leaves their relative chronology indeterminable by direct means. The Long Loch and Priomh Loch tongues are pre-emplacement of the solidified E.L.S. (cut by the Inner Ring Fault), and may therefore represent mobilized unconsolidated olivine cumulates from the E.L.S. magma chamber. If so, the absence of harrisitic (skeletal) olivines is surprising, considering the abundance of harrisite layers within the exposed section of the E.L.S. (Chapter 2).

The extreme adcumulate texture of the dunite plugs (Fo_{87}-Fo_{91}) is not regarded as the result of modal changes due to postcumulus metasomatism by filtering interstitial silicate liquid, as postulated

for several "replacement" dunite plugs in ultramafites from the Stillwater, Bushveld and Duke Island complexes (Hess, 1960; Cameron and Desborough, 1964; Irvine, 1974, 1980b). The process of adcumulus growth may occur deep within a cumulate pile principally as a result of simple compaction and continued growth from the liquid in response to local thermal gradients (Irvine, 1980b). However, some additional process or processes are required in order to form the more extreme adcumulates of the dunite plugs:

(i) a limited amount of the "extra" components of the residual intercumulus liquid may be incorporated into solid solution in the cumulus phases (olivine and spinel) e.g. Al_2O_3 and some TiO_2 into the Cr-spinel;

(ii) the remaining components (e.g. Na_2O, K_2O, P_2O_5) may have been eliminated by diffusion through the intercumulus liquid. Heat loss through the margins of these dunitic (and other peridotitic) bodies was minimal, as indicated by the absence of chilling effects in the intercumulus material: the host layered series rocks may have been heated (possibly over a long period of time) by relatively large volumes of liquid, or liquid plus crystals, moving through the fracture prior to filling by the highly-porphyritic magma. As a result of limited heat transfer from plug to host rocks, postcumulus crystallization was sufficiently slow (Irvine, 1970b, 1974b) to allow ample time for intercumulus diffusion through distances of a few tens of meters.

The black peridotites may be the youngest of the ultramafic intrusions, by virtue of (a) their relatively fresh nature, and (b) their more evolved olivine compositions (Fo_{83}).

The relative ages of the plugs inside (e.g. E107, E65, B29) and outside (M50, M42, M32) the L.U.C. is indeterminable: they may be

"early" (contemporaneous with the tongues) or "late" (contemporaneous with the Central Series).

CHAPTER 6

GEOCHEMISTRY AND PETROGENESIS

6.1 Introduction

Some 53 samples (20 dykes, 13 gabbros and 20 ultrabasic rocks) have been analysed for all major and 17 trace elements. Analytical procedures are described in Appendix B, and analyses and CIPW norms listed in Table 6.1 and 6.2. The precision and accuracy of the X.R.F. data are summarized in Tables B.3 and B.4.

In Chapter4 the dykes were divided into three petrographic groups A, B and C: this does not imply separate origins for the three groups - they may well constitute a single trend. The gabbros (sampled from Groups 1, 2, 3, 5) will be treated as two groups (a) one pyroxene (cpx), and (b) two pyroxene (cpx-opx). The ultrabasic rocks include the allivalites, layered peridotites, and non-layered peridotitic minor intrusions.

6.2 Classification

The nature and diversity of igneous activity within the British Tertiary Province is well-documented, and in order to avoid ambiguity a division into rock types and magma types, based on major and trace element data, needs special care. Three distinct basaltic "magma types" (showing internal chemical variation, but not related to each other by low-pressure crystal-liquid processes) have been recognized on Skye (Mattey *et al.*, 1977); (i) the Skye Main Lava Series, SMLS (transitional basalts), (ii) the Preshal Mhor type (low-K tholeiites) and (iii) the Fairy Bridge type (transitional). Equivalents of (i) and (ii) have been noted in NE Ireland (Gamble, 1979), Mull (Lamacraft, 1977) and Rhum (Forster, 1980), and it has been suggested (Mattey *et al.*, 1977; Morrison, 1978) that other Tertiary igneous centres may contain a range of basalt magma types similar to that observed on Skye.

6.2.1 Effects of alteration

Prior to classification, and in order to discuss magmatic geochemical variation, adjustments must be made for post-solidification changes which may have significantly altered the rocks' original chemistry. Several alteration processes have been recognized, which can operate simultaneously or independently (Morrison, 1978).

(i) weathering

(ii) zeolitization, attributed to circulating alkaline solutions

(iii) hydrothermal alteration, attributed to convective circulation of groundwater.

(i) Collection of only fresh material where possible, and the removal of weathered surfaces during sample preparation, will minimize the effects of weathering.

(ii) Amygdaloidal and interstitial zeolites are rare; zeolitization is therefore not considered to have significantly altered the geochemistry of the samples.

(iii) The majority of the Rhum dykes have suffered only slight hydrothermal alteration; they show incipient breakdown of groundmass mafic minerals to serpentine, chlorite and iron oxides, but albitization of plagioclase is rare. The gabbros are slightly more altered, with serpentinized olivine and chloritized clinopyroxene. Serpentinization is maximum (ca. 5-10%) in the non-layered peridotites. Studies of element mobility in altered basaltic rocks (Floyd and Winchester, 1975; Wood *et al.*, 1976; Morrison, 1978; Humphris *et al*, 1978) have indicated that only Al_2O_3, TiO_2, P_2O_5, Nb, Cr, Zr, and Y are considered to have been immobile during alteration. Their stability is greater in transitional and alkaline basalts than in tholeiites (Humphris *et al.*, 1978; Morrison, 1978). Contents of all other elements, especially alkalis, Rb, Sr, and Ba, may be suspect.

Post-intrusion oxidation can significantly alter the Fe_2O_3/FeO ratio from the magmatic value (Irvine and Barager, 1971; Thompson *et al.*, 1972; Brooks, 1976; Clarke and O'Hara, 1979), and hence appreciably affect (a) the proportion of *ne* or *hy* in the CIPW normative calculations (Coombs, 1963), and (b) subsequent classification of the rocks. Several methods of adjustment have been proposed: wt% Fe_2O_3 = wt% TiO_2 + 1.5 (Irvine and Barager, 1971); wt% Fe_2O_3 = 1.5 if $Na_2O + K_2O < 4.0$ wt% (Thompson *et al.*, 1972); $Fe_2O_3/FeO = 0.15$ (Brooks, 1976); $Fe_2O_3/FeO+Fe_2O_3 = 0.15$ (Clarke and O'Hara, 1979). An $Fe_2O_3/Fe_2O_3 + FeO$ ratio of 0.18 has been chosen, based on an average value obtained by wet chemical method for less altered gabbros of similar bulk composition (Brown, 1956).

6.2.2 Geochemical classification parameters

Classification and nomenclature of the dykes are based on the following parameters:

(1) MgO wt%: MgO is used to distinguish between the basaltic (<9% MgO) and picritic or high-MgO (>9% MgO) suites.

(2) normative mineralogy: including D.I. (Thornton and Tuttle, 1960), abundance of normative *ne, hy, ol,* and normative composition of the feldspar.

(3) total wt% alkalis vs wt% SiO_2.

6.2.3 Classification

In terms of total alkalis and silica (fig 6.2) the dykes and 1-pyx gabbros are transitional, straddling the Hawaiian discriminant line dividing the tholeiitic and alkali olivine basalt fields (MacDonald and Katsura, 1964). The 2-pyx gabbros plot within the tholeiitic field.

Using the classification scheme presented in fig 6.3 the majority of the dykes and all the gabbros are "basaltic" (normative plagioclase

$>An_{50}$); two dykes are intermediate in composition (M189 falls in the basaltic hawaiite category and M59/A lies just within the mugearite field). The various basaltic groups and subgroups define separate clusters of data, and with differentiation form the sequence A-3, A-1, A-2, B, C.

The majority of the dykes and gabbros contain significant amounts of both normative *ol* and *hy* (>8.3 and >1.5%, respectively, Table 6.1) and fall into the olivine tholeiite division of Yoder and Tilley (1962). In view of the selection of only the freshest material for analysis, the normative *hy* is considered "primary". However their classification as tholeiites is at variance with their petrographic similarity to alkali basalts, and they are best termed mildly alkaline or transitional basalts (Bass, 1972). Those with alkaline affinities have olivine in the groundmass (Group A and one sample from Group B); those with tholeiitic affinities (the majority of Group B) lack groundmass olivine. Nine dykes (from Groups A, B and C) have minor amounts of normative *ne* (Table 6.1 and fig 6.4).

Differences between the various groups in terms of silica-saturation are slight (fig 6.4); both dykes and gabbros straddle the "critical plane of silica undersaturation". With increasing D.I. the dykes show an overall progressive change from mildly silica-saturated to mildly silica-undersaturated (*ne*-normative) compositions. Similar trends are defined by individual groups (A and B). D.I.'s range from 5.6-18.6 (Group A); 16.1-25.8 (Group B); to 39.5 (Group C). The picritic and basaltic dykes both have mildly *ne*- and *hy*-normative members. However, the two groups are characterized by differing proportions of a) normative olivine: >20% (picritic) and 8.3-13.2% (basaltic), and b) normative plagioclase composition: $>An_{60}$ in the picrites and generally $<An_{60}$ in the basalts (fig 6.3).

The two gabbro groups have similar ranges of D.I.: 11.0-23.6 (1-pyx) and 13.1-24.0 (2-pyx). With increasing D.I. the 1-pyx group shows a tendency towards Si-undersaturation; the 2-pyx gabbros show slight enrichment in "Q" (fig 6.4).

Transitional basalts are a widespread product of early major Tertiary volcanicity in the Scottish province: the Skye Main Lava Series (Thompson *et al.*, 1972) and Fairy Bridge magma type (Mattey *et al.*, 1977) of Skye; the Mull Plateau Group (Morrison, 1978); the Small Isles volcanics (Ridley, 1971, 1973) and the majority of dykes from the Rhum dyke swarm (Forster, 1980) all contain both *ne*- and *hy*-normative varieties.

The petrography (olivine, plagioclase and clinopyroxene phenocrysts) and major element composition (see section 6.3) of the Rhum transitional basaltic rocks suggest that they approach low-pressure ol-plag-cpx-liquid cotectic equilibria in the natural basalt system.

6.3 Major Element Geochemistry

Oxide vs MgO (Bowen) variation diagrams (fig 6.1) effectively portray the chemical variations. Fractionation trends show clearly the effects of the removal of olivine, spinel, plagioclase and clinopyroxene from high-MgO basalt liquids. Approach to a liquid line of descent is dependent on the phenocryst-content of the samples: Group B dykes (<9 wt% MgO) are aphyric (<5% phenocrysts) and clearly approximate to liquid compositions, whilst Group A picritic dykes are generally enriched in olivine phenocrysts (up to 60%), and the gabbros were intruded as liquids carrying variable (but small, <5%) contents of olivine, plagioclase and clinopyroxene crystals (Chapter 3).

The AFM variation diagram (fig 6.5) demonstrates moderate iron-enrichment and a close approximation to trends recorded from other transitional and tholeiitic suites.

6.3.1 Dykes and gabbros

Major element data (fig 6.1) show a reasonably coherent serial variation from picrites (33.7-31.0, 25.8-17.0 and 16.1-9.3 wt% MgO for groups A-3,A-1,A-2 respectively) to basalts (9.1-6.5% MgO) to the "differentiated" (i.e. MgO <6.5%) Group C dykes with 4.8% MgO. Gabbros have a similar range of MgO-contents (20.3-7.7%). Chemical characteristics are reflected in the phenocryst mineralogies (summarized in fig 6.1); differentiation in the picrites is controlled by olivine (and spinel) fractionation, and in basalts and gabbros by olivine, plagioclase and clinopyroxene. A mass balance calculation indicates that the separation of ca. 33% olivine (av. Fo_{88}) from the most basic picrite (M9, 20.5% MgO) would yield a residual liquid of the composition of the most primitive basalt (M184, 9.3% MgO).

In the plot of total iron (as Fe_2O_3) the fractionation sequence is especially clear. Samples with >14% MgO have olivine (plus minor spinel) as the sole phenocryst and lie on a fractionation curve from $Fo_{92.5}$ (the most primitive phenocryst composition). The sudden drop in total iron at MgO-levels <14% corresponds to the appearance of liquidus Al-Cr-magnetite (Chapter 4, section 4.4.4.1). A second inflection (an increase) at ca. 9% MgO, corresponds to the appearance of liquidus plagioclase closely followed by clinopyroxene.

Gabbro data display a scatter (due to varying degrees of cumulus enrichment) but generally define a trend controlled by removal of plagioclase and a less-magnesian olivine (Fo_{77}) in the ratio 1:2. As in the dykes, the appearance of clinopyroxene on the cotectic occurs at ca. 10% MgO. The lower Fe_2O_3-contents of the gabbros (relative to the dykes) for a given MgO-content may be due to the "dilution effect" of accumulative plagioclase crystals.

Al_2O_3 data (fig 6.1) confirm the co-crystallization of minor amounts of Al-rich spinel with the magnesian olivine in the picritic dykes. Samples on the MgO-poor side of the Al_2O_3 maximum, as expected, contain plagioclase and clinopyroxene phenocrysts in addition to olivine. The spread of Al_2O_3 and CaO data for the gabbros is due to varying cumulus plagioclase contents.

MnO data show considerable scatter for a given MgO-content, reflecting varying opaque oxide contents. After an initial increase with declining MgO, MnO-levels in the dykes remain approximately constant due to the higher MnO-contents of the coexisting olivines. The positive correlation of MnO with MgO, in the gabbros, is a consequence of previous MnO-depletion by olivine fractionation.

TiO_2 in the dykes (fig 6.1) shows a general increase with fractionation. The *ne*-normative basalts tend to have higher TiO_2-contents than *hy*-normative samples. The gabbros are depleted in TiO_2 relative to the dykes.

P_2O_5, Na_2O and K_2O show trends typical for fractionated basalt series, with concentration in the late liquids. P_2O_5 and K_2O data scatter is suggestive of involvement of these elements in alteration processes. P_2O_5, like TiO_2, is enriched in *ne*-normative relative to *hy*-normative basalts. No such difference can be discerned for the K_2O data.

6.3.2 The ultrabasic rocks

As expected, analyses of the olivine and plagioclase cumulates define a line joining their "average" plagioclase and olivine compositions (An_{88} and Fo_{84} respectively). The degree of deviation from this line is directly proportional to the content of intercumulus material, mainly plagioclase and clinopyroxene.

6.3.3 Summary

The major element compositions of the picrite → mugearite dyke suite can be approximated to a liquid line of descent controlled by fractionation of the observed phenocrysts (olivine, Al-rich spinel, Cr-,Fe-rich spinel, plagioclase and clinopyroxene). The dry, 1 atmosphere experimental liquidus of chilled, aphyric sample B65 (13.5% MgO) lies close to 1320 ± 5°C, with olivine on the liquidus (J. Craven pers. comm.). Below ca. 1275°C olivine and plagioclase coprecipitate, and the ol-plag-cpx-liq (1 atm) cotectic is attained at ca. 1206°C.

The major element variation in the gabbros is the result of fractionation of olivine and plagioclase in approximately a 2.5:1 ratio (this fits with the proposed petrographic relationship to the Eastern Layered Series (E.L.S.): the E.L.S. has a ratio of olivine cumulates (peridotite) to plagioclase cumulates (allivalite) of ca. 3:1).

The dyke and gabbro liquids may be related by addition/subtraction of the observed phenocryst phases: apparent differences in TiO_2-, MnO-, Al_2O_3- and Fe_2O_3-contents are attributable to accumulative plagioclase "dilution" in the gabbros.

6.3.4 Comparison with other U.K. Tertiary magmas

Major oxide variations of the Rhum dykes and gabbros (this study) are compared with those of the transitional basalt suites of the Small Isles, Skye and Mull (fig 6.6). Additional basaltic magma types recognized on Skye (Preshal Mhor and Fairy Bridge types, Mattey *et al.*, 1977) and Mull (Group I-III tholeiitic lavas, Beckinsale *et al.*, 1978) are included. The Rhum dykes have very similar major element chemistries to the Skye Main Lava Series, Mull Plateau Group and Fairy Bridge type transitional basalts, but have higher CaO-(10.5-13.5%) and lower Na_2O-(1.5-3.2%), K_2O-(0.01-0.4%) and P_2O_5-(0.01-0.18%)

contents, more typical of the Preshal Mhor type. The high CaO-levels cannot be attributed to plagioclase enrichment (e.g. in the picrite dykes). The gabbros appear closely comparable to the Preshal Mhor magma type in all major element contents, but part, if not all, of this apparent similarity may be the product of varying degrees of cumulus plagioclase-enrichment.

6.4 Trace Element Chemistry

The absolute and relative abundances of compatible (distribution coefficient D >1) and incompatible (D <1) trace elements can carry significant information bearing on the genetic relations of basaltic liquids. Rocks with broadly similar major element compositions may be widely variable in terms of incompatible trace elements. Trace element concentrations are governed by (i) concentration in the source rocks, (ii) extent of fractional melting, (iii) extent of fractionation during ascent and crystallization of the liquid, (iv) wall-rock reactions, (v) magma mixing. Meaningful interpretation is restricted by the paucity of data on trace element partitioning between the mafic phases (especially ol and cpx) and liquid. Allowing for extension of the range of trace element concentrations by crystal accumulation, variation in trace element chemistry within a suite of supposedly related rocks may be (a) fitted to a single crystal-fractionation model for a common parent magma, or (b) ascribed to contamination or source region heterogeneity.

Analyses for 3 light rare-earth elements (LREE) and 14 other trace elements are reported: the data are used to test and amplify the evolutionary model for Rhum, proposed in the preceding major element section. Trace element analyses (listed in Table 6.2) are presented as ppm vs wt% MgO variation diagrams in figs 6.7-6.16.

Variation in the transition elements, e.g. Ni, Cr, is in keeping with their compatible character: they partition into the phases demonstrably involved in fractionation of the Rhum magmas. Ni (fig 6.7) is rapidly depleted (to 65 ppm at <6.5% MgO) by the fractionation of olivine and minor sulphides. The decline in Cr is due to removal of Cr-spinel and later, clinopyroxene; scatter and extension of ranges of these elements (Ni 2038-65 ppm, Cr 2380-62 ppm) are due to variable enrichment in olivine and spinel phenocrysts. Ni-concentrations are comparable to those predicted by Duke (1979) for a model komatiitic system with sulphide fractionation and a 1000:1 ratio of separating olivine and sulphide; NiO-contents of the olivines (Chapter 4) confirm the Rhum trend is indeed sulphur-saturated. MgO-Ni correlation is discussed in more detail in section 6.5.1, with regard to the recognition of primitive high-MgO liquids.

A mass balance calculation indicates that separation of ca. 30% olivine (Fo_{92}, 0.40 wt% NiO) from the most basic picritic liquid (M9 groundmass, 20.5 wt% MgO, 950 ppm Ni) would yield a residual liquid with the Ni composition of the most basic basalt (M184, 9.3% MgO, 149 ppm Ni).

Since Ni is depleted more rapidly than Cr, the Cr/Ni ratio of the liquid increases with differentiation (Table 6.3). The three groups of picrites can be distinguished according to their Cr/Ni ratio (A-3, <1.27; A-2, av. 1.5; A-1, 1.4-2.9); the 1-pyx and 2-pyx gabbros cannot. Group B basaltic dykes have similar Cr/Ni ratios to the A-2 picrites (1.5).

Vanadium and scandium (fig 6.8) show a progressive increase with fractionation in both dykes and gabbros. Both elements partition into clinopyroxene and Fe-Ti oxides (Taylor *et al.*, 1969; Ewart *et al.*, 1973), therefore data scatter is interpreted in terms of varying modal

contents of these phases. For a given MgO-value, V- and Sc-contents are consistently lower in the gabbros.

Copper (fig 6.9) shows a progressive increase with fractionation, to a peak at ca. 10% MgO, and thereafter decreases rapidly during plagioclase and clinopyroxene fractionation. The Cu trend is crudely comparable to the Al_2O_3 and CaO trends; Cu may enter Na^+ and Ca^{2+} positions in plagioclase and Al^{3+} and Fe^{2+} positions in clinopyroxene (Taylor, 1965). Alternatively the trend can be interpreted in terms of Cu-sulphide fractionation. The initial Cu-enrichment is characteristic of sulphur-saturated fractionation when the olivine/Ni-sulphide ratio is >175 (Duke, 1979 figs 1 and 4). At a critical concentration (ca. 180 ppm Cu at 10% MgO) Cu-bearing sulphides separate, and Cu is rapidly depleted in the liquid, (chalcopyrite and bornite are the dominant sulphide phases in gabbros and dykes with <10% MgO).

Cu-contents are significantly higher than those presented by Duke (1979) for fractionation of sulphide-saturated komatiitic and mid-ocean ridge basalts (fig 6.9), and show a lower rate of increase with declining MgO. These features may be due to (a) higher initial Cu-concentrations in the Rhum parent liquids, or (b) a partition coefficient for Cu between sulphide and silicate liquids, lower than the 178 value used by Duke and Naldrett (1978). The Ni/Cu ratio of the differentiating Rhum liquid decreases steadily (32.7-0.21, fig 6.10), then increases slightly in the late stages, as Cu-sulphides fractionate. Values are lower than those predicted by Duke (1979) because of the higher Cu-contents in the Rhum samples.

Zn (fig 6.11) substitutes for Fe^{2+} (Taylor, 1965) and concentrates into magnetite. A slight decrease in Zn (greater in the gabbros) in the middle stages of differentiation suggests fractionation of Zn by Fe-Cu-Ni sulphides. Pb (fig 6.11) shows no discernible trend.

Contents of Ba, Rb and Sr (<110, <5 and <370 ppm respectively, fig 6.12) are low: like the Hebridean Tertiary basalts of Skye and Mull (Morrison *et al.*, 1980), the Rhum samples are depleted in these elements (by factors of up to 10) relative to other mildly alkaline to transitional basalts.

K and Rb show a positive correlation (fig 6.13) in all rock types; K/Rb ratios (fig 6.14 and Table 6.3) show maximum variation in the gabbros, (high ratios >700 in some dykes are the result of alteration). Ratios in the gabbros increase with differentiation (275-538 and 220-250 in 1-pyx and 2-pyx groups respectively) due to separation of plagioclase >An_{75}, with a K/Rb distribution coefficient of <1 (Philpotts and Schnetzler, 1970). This supports the hypothesis for a genetic relationship between the gabbros and L.U.C.; the former represent near-liquid compositions which have previously fractionated olivine and calcic plagioclase (>An_{75}), while the latter represent the separated phases. K/Rb ratios in the basaltic dykes decrease (718-293) due to the separation of plagioclase (<An_{75}) with higher K/Rb ratios than their equilibrium liquids (Philpotts and Schnetzler, 1970).

Similar patterns and conclusions are deduced from K/Ba ratios (Table 6.3).

Sr-levels (fig 6.12) in the basaltic dykes decrease with fractionation (373-225 ppm) due to separation of plagioclase (D_{Sr}^{pl-liq}=1.3-2.8, Philpotts and Schnetzler, 1970). Similarly plagioclase fractionation was important in the genesis of the gabbros (344-186 ppm Sr).

Nb-and Th-concentrations are low (fig 6.15); data show considerable scatter and no discernible trends. With its similar ionic radius to Ti, Nb would be expected to increase in the liquid prior to fractionation of a Ti-rich phase.

Zr and Y increase steadily with differentiation (42-427 and 10-73 ppm respectively, fig 6.16): concentrations are significantly lower in the gabbros (8-132 ppm Zr and 6-24 ppm Y). Ti/Zr and Y/Zr ratios remain approximately constant (Table 6.3), averaging 120 and 0.15 respectively (fig 6.17). The gabbros display a greater range of both ratios.

6.4.1 Comparison with other Hebridean Tertiary magmas

Trace element variations (excluding the rare earth elements, see section 6.4.2) are compared in Table 6.4 with those of the Small Isles, Skye, and Mull basalt suites. Published data for the transition elements are few. Zr, Y, V, Nb and Pb concentrations are similar throughout the suites; the Rhum dykes are depleted in Rb, Ba and Sr.

Ti/Zr ratios (which should reflect those in the primary liquid, and hence those of the source rocks) average 120 in both transitional (Rhum, Mull) and tholeiitic (Preshal Mhor) basalts (fig 6.17); those in the Skye Main Lava Series are distinctly lower and show a wider range.

High Y/Zr ratios (0.5) distinguish the Preshal Mhor and Fairy Bridge types from Rhum and other Hebridean basalts; therefore neither magma type is represented in the dyke swarm cutting the Rhum L.U.C. Preshal Mhor dykes do cut the Rhum lava pile (Forster, 1980) but may be outliers from the Skye swarm.

6.4.2 Rare earth element geochemistry

Chondrite-normalized light rare earth element (LREE) data are presented in fig 6.18. Absolute La, Ce and Nd abundances are listed in Table 6.2. Cumulates have LREE-contents below detection limits, due to low LREE distribution coefficients (Schnetzler and Philpotts, 1970). The main features of fig 6.18 are:

(i) abundances (chondrite-normalized) increase by an order of magnitude

in the sequence gabbros-picrite-basalt-intermediate dykes, and are higher in *ne*-normative, relative to *hy*-normative samples.

(ii) slopes (expressed as $(La/Nd)_N$, Table 6.3) range 0.5-1.6 in the dykes and 0.5-2.4 in the gabbros, with no difference between *ne*- and *hy*-normative types.

(iii) curvatures (expressed as $(Ce-La)_N/Nd_N$, Table 6.3) show no correlation with Si-saturation.

(iv) Y behaves geochemically much as the heavy REE Er, and normalized abundances (fig 6.18) suggest a LREE-enriched pattern for both gabbros and dykes.

The LREE patterns for cumulus plagioclase in the L.U.C. (Henderson and Gijbels, 1976) support the hypothesis of a genetic link between the gabbros and L.U.C.

Rhum's LREE-enriched pattern is distinct from the LREE-depleted pattern of the Preshal Mhor magma type (fig 6.19); it more closely resembles those of the Skye Main Lava Series (SMLS), Mull Plateau Group (MPG) and Fairy Bridge basalts. Similar REE variations are reported from magnesian basalts from the FAMOUS area of the Mid-Atlantic Ridge (Langmuir *et al.*, 1977). The significance of the REE data is discussed in more detail in sections 6.5.3 and 6.5.4.

6.5 Petrogenesis

The major and trace element variation in the basaltic minor intrusions may be ascribed to one or more of the following:

(a) primary variation established at the site of magma generation, due to e.g. varying degree of melting; type of melting (e.g. equilibrium or fractional); source rock heterogeneity; variable P_{O_2}, P_{Co_2} etc. in source regions; (b) polybaric fractional crystallization; (c) crustal contamination; (d) magma mixing.

The compositional diversification is interpreted in terms of low-pressure crystal fractionation of the observed phenocryst phases (the "first-order" model). This low-pressure variation may be the result of a complex sequence of polybaric events at <10 kb, e.g. opportunities for fractionation are a function of (i) rate of primary liquid generation and its intrusion into the crust, (ii) volume of individual batches, (iii) rate of ascent through the crust, (iv) development of magma chambers at one or more crustal levels, and (v) residence times within chamber(s).

However, the smallness of the dykes (<1 m wide) and the very limited differentiation shown, suggest that the dyke liquids fractionated solely during ascent from lower crustal or upper mantle "holding chambers", and may not have been expelled as residual liquids from a higher level (upper crustal) chamber.

6.5.1 Parental liquid composition and the existence of high-MgO liquids in Rhum

Criteria for the recognition of "liquid compositions" amongst porphyritic basalts have been the cause of considerable controversy in recent years, especially with regard to the existence of high-MgO (>9% MgO) liquids (Cox, 1978; Clarke and O'Hara, 1979).

Argument centres around two extreme points of view; (1) that the commonly-erupted basalts (<9% MgO) are primary, i.e. unmodified from their site of generation, and the more picritic related compositions are cumulus-enriched; and (2) that high-MgO liquids do exist in nature, and are parental to most of the observed basalts; the latter are derivatives through a process of olivine removal.

On the basis of field, petrographic, geochemical and experimental studies, many workers now firmly believe in high-MgO liquids (Drever and Johnston, 1957; Clarke, 1970; Cox, 1978, 1980; Jakobsson *et al.*,

1978; Malpas, 1978; Elthon, 1979; Clarke and O'Hara, 1979; Maaloe, 1979; Maaloe and Jakobsson, 1980). Acceptance was hastened by the discovery of komatiites with MgO-contents >30% (Viljoen and Viljoen, 1969a, b), and the recognition that the spinifex-textures resulted from rapid crystallization of liquids with similar bulk compositions (Nesbitt, 1971; Bickle *et al.*, 1975).

Several methods can be used to distinguish possible parental and derived liquid compositions.

(i) study of Fe-Mg distribution between olivine and coexisting liquids. Under equilibrium conditions the Fe-Mg K_D for olivine is 0.3 ± 0.03 (Roeder and Emslie, 1970; Longhi *et al.*, 1978); a slightly higher value (0.33) is predicted for high-MgO liquids (Cawthorn *et al.*, 1974; O'Hara, 1977). Higher values (>0.33) imply an origin by olivine accumulation (Flower *et al.*, 1977). Calculated Fe-Mg K_D^{ol-liq} values for the Rhum picrites (Table 6.5) suggest that the dykes (with the exception of B62/2) contain some accumulated olivine. Olivine-enrichment (also suggested by the discrepancy between calculated liquidus olivine compositions and observed phenocryst cores, Table 6.5) may be the result of olivine redistribution, e.g. flow differentiation.

(ii) Ni-Mg distribution between olivine and coexisting liquid has been widely used as a criterion for identifying primary magmas and liquids (Sato, 1977; Arndt, 1977; Hart and Davis, 1978; Clarke and O'Hara, 1979). Equations relating D_{Ni}^{ol-liq} to the MgO-content of coexisting liquid (Arndt, 1977; Hart and Davis, 1978) can be used to model the MgO-Ni fractionation paths within rock suites. Ni-MgO data for the Rhum dykes (fig 6.20) show a reasonably good fit to the batch or Rayleigh fractionation trends of Arndt, and are closely comparable to those of the Baffin picrite-basalt suite. The lower than predicted

Ni-levels in liquids with <12% MgO are consistent with the hypothesis for fractionation involving an increasing proportion of sulphides (Duke, 1979); sulphides are modally more abundant in the basalts than the picrites.

(iii) selection of the most primitive parental liquid composition on Rhum is based on the most basic (MgO-rich) of (a) the aphyric picrites, or (b) the picrite groundmass compositions. The aphyric picrite B65 contains 13.47 wt% MgO (Table 6.1), but the quenched groundmass in M9 has 20.5% MgO (Table 6.6). In view of the known efficiency of gravitative fractionation of olivine at near-liquidus temperatures, and the expansion of the stability field for olivine with decreasing pressure, it is highly probable that the dyke liquids evolved during ascent by olivine fractionation from more Mg-rich precursors. The figure of 20.5% may therefore represent a minimum MgO-content of the primary basaltic liquid.

Liquids with comparable MgO-contents (Table 6.6) have been inferred as parental to oceanic crust (Elthon, 1979), continental flood basalts (Deccan, Krishnamurthy and Cox, 1977; Lebombo and Nuanetsi, Bristow, 1980) and mid-ocean ridge basalts (Baffin Bay, Clarke, 1970; Reykjanes Peninsula, Jakobsson *et al.*, 1978). The Rhum liquids are poorer in SiO_2, richer in TiO_2, but otherwise very similar to those from Baffin Bay (compare analyses 2 and 9). They are strongly depleted in SiO_2, TiO_2, CaO, alkalis, P_2O_5 and all trace elements except Cu, Cr, V and Sc, relative to picritic basalts from continental flood provinces (compare analyses 2 and 14), but slightly richer in Al_2O_3, alkalis and TiO_2, than Tertiary komatiites (compare analyses 2 and 25-26).

Despite field, petrological, geochemical and experimental evidence for the existence of picritic (MgO >9 wt%) primary liquids, natural

examples of such liquids are rare. For example <10% of published analyses of mid-ocean ridge basalt glass compositions (Bryan and Moore, 1977; Bryan, 1979) contain >9% MgO; the most primitive glasses yet observed (Bryan and Moore, 1977) have only 10-12% MgO. In the continental environment a fundamental constraint on the ascent (and eruption) of picritic liquids is their high density relative to that of average continental crust (>2.73 g/cm^3 for liquids with >18.5% MgO: the occurrence of phenocrysts increases density, Huppert and Sparks, 1980a, b). In effect, the crust imposes a density filter on liquids passing through it (the "window of eruptibility" concept of Stolper and Walker, 1980). Fluid dynamical models (Sparks *et al.*, 1980; Stolper and Walker, 1980; Huppert and Sparks, 1980a, b) predict that high-MgO liquids are "ponded" at the base of the crust, where they undergo fractionation of predominantly olivine to form ultramafic cumulates. Such a process may have occurred beneath the Hebridean crust in the Rhum instance.

With the separation of abundant olivine, density of the liquid decreases until plag ± cpx join olivine as the crystallizing phase; density then increases (Sparks *et al.*, 1980). Liquids which occupy this minimum (9-11% MgO) *can* rise through the oceanic or continental crust: significantly these are the compositions of the most primitive mid-ocean ridge glasses (Sparks *et al.*, 1980).

6.5.2 Origin of high-MgO liquids

Primary liquid formed by partial melting of upper mantle, will form in equilibrium with olivine ± orthopyroxene and clinopyroxene (O'Hara, 1968a), and it is relevant to consider the conditions under which such liquid compositions could be generated by dry melting of mantle material.

The upper mantle lherzolite contains garnet or spinel at most pressures and temperatures up to the solidus (except at lowest pressures

where a plagioclase lherzolite stability field may intervene), O'Hara (1968a): garnet lherzolite is regarded as the source of basaltic liquids (Yoder, 1976, Chapter 2).

Assuming high-MgO liquids are mantle melts, their composition will be controlled by (i) the composition and mineralogy of the source material, (ii) the degree of melting, (iii) the nature of the residue, (iv) P-T conditions at the point of separation of the melt, (v) crystal fractionation.

The transitional nature of the Rhum dykes (Table 6.1), together with the transitional volcanics and intrusives from the Small Isles (Ridley, 1971, 1973), Ardnamurchan (Holland and Brown, 1971), Skye (Thompson *et al.*, 1972, 1974a), Mull (Beckinsale *et al.*, 1978) and other worldwide transitional basalts (Green, 1970; Bass, 1972) reflects derivation at such depths that the critical plane of silica-undersaturation no longer exists as a "thermal divide". Estimates of the depth extent of the thermal divide range from 8 kb (O'Hara, 1968a) to 3-4 kb (Presnall *et al.*, 1978): the former is generally preferred, therefore transitional liquids originate from depths >25 km. Recent geophysical work estimates the Hebridean crust to be ca. 27 km thick (Bott *et al.*, 1979). Consequently the thermal divide would not be operative in the lower crust or upper mantle beneath Rhum, and the coexistence of *ne*- and *hy*-normative dyke compositions must reflect one or more of the following:

(a) simultaneous differing degrees of partial melting; (b) mantle heterogeneity; (c) simultaneous differing depths of melting (d) repeated crossing of the critical plane of silica undersaturation (since at these depths it no longer exists as a thermal divide) as a result of crystal fractionation; (e) crustal contamination.

(a) Schilling and Winchester (1967) theoretically analysed the sequence of REE changes with degree of melting, and although the

calculated curves were not directly correlated with the normative character of the deduced liquids, in Schilling's view the REE data constituted the strongest argument that the degree of melting influences the normative character of the melt; such that it changes from *ne* to *hy* as the degree of melting increases (at constant pressures). However, no single bulk composition has yet been shown to produce such a series of liquids over a moderate range of melting (ca. 30%).

(b) The coexistence of *ne*- and *hy*-normative basalts with the same REE pattern (Schilling and Bonatti, 1975) casts further doubt on the principal role of degree of melting in controlling the Si-saturation of liquids produced: the two magma types may be products of approximately the same degree of melting, but at different eutectic compositions.

(c) Experimental studies of basalt systems (Green and Ringwood, 1967; O'Hara, 1965; Kushiro, 1973) have shown that the normative character of initial melts (in equilibrium with a four phase lherzolite) varies with increasing pressure from *q*- (<5 kb) to *hy*- (5-8 kb) to *ne*- (8-20 kb) to *hy*- (20-40 kb). More advanced melting yields *hy*-normative liquids at all pressures. Consequently the narrow range of normative *ne* and *hy* contents in the Rhum dykes may result from similar degrees of melting occurring simultaneously throughout a wide pressure (depth) range.

(d) alternatively, fractionation of garnet (Yoder and Tilley, 1962), subcalcic clinopyroxene (Tilley and Yoder, 1964), orthopyroxene (Green and Ringwood, 1967) or all three (O'Hara, 1965) can produce a range of Si-saturated and undersaturated liquids from homogeneous *hy*- or *ne*-normative primary melts.

(e) Morrison *et al.* (1980) rejected, whilst Wood (1980) supported, a hypothesis that the range of Si-saturation in the Skye and Mull basalts resulted from varying degrees of contamination of chemically-

uniform alkalic magmas by lower and upper crustal gneisses (granulite and amphibolite facies respectively).

Considering the wealth of recently-published data concerning the Hebridean basalts (Mattey *et al.*, 1977; Beckinsale *et al.*, 1978; Thompson *et al.*, 1980; Morrison *et al.*, 1980), the general consensus is that their range of Si-saturation probably results from a combination of varying degrees and depths of melting.

In the absence of experimental data and significant geochemical constraints (e.g. reliable REE patterns), estimates of the source depths and degree of melting for generation of the Rhum primary liquids are purely speculative.

Three suggestions have been made as to the depths at which picritic (MgO >9 wt%) primary liquids can be generated (reviewed by Stolper, 1980): (i) at 25-30 kb, leaving ol-opx-cpx-gt as residual phases (O'Hara, 1968a); (ii) at 20 kb, leaving only ol-opx ± spinel in the residue (Green *et al.*, 1979); (iii) at 5-10 kb, with ol-opx as residual phases (Elthon, 1979). Stolper's (1980) data preclude (iii), favour (ii), but cannot rule out (i).

With regard to the degree of melting involved in the generation of liquids with 20% MgO, a figure of ca. 10-15% is suggested by O'Hara (1968a), and Maaloe and Jakobsson (1980): a similar figure (20%) has been suggested for Baffin Group II olivine basalts (Elthon and Ridley, 1979). Similarity of the Baffin basalts (20% MgO) to the most primitive Rhum picrite (20.5% MgO) has already been noted (Table 6.6), and it could be argued that the "transitional" nature of the Rhum liquids points to a deeper source than for Baffin.

6.5.3 Trace element constraints on magma genesis

Trace elements can be helpful in specifying conditions of melt generation and the nature of the source material (Hart and Allegre,

1980). The relative constancy of the incompatible element ratios e.g. Ti/Zr, Y/Zr, Zr/Nb (Table 6.3) suggests a constant degree of melting was involved (these ratios are unaffected by olivine fractionation). The ratios are close to chondritic values and approximate to source mantle ratios (assuming chondritic abundances in the upper mantle). 20-25% melting is envisaged in the Hebridean mantle: smaller degrees result in lower ratios, since Zr would be preferentially enriched in the melt (supporting evidence is the decrease in e.g. Y/Zr from tholeiitic to alkali basalt, Erlank and Kable, 1976).

Chondrite-normalized REE patterns are important indicators of petrogenetic processes (Yoder, 1976, Chapter 8): the flat or slightly LREE-enriched patterns for the Rhum basalts (fig 6.19) are suggestive of slight to moderate degrees of melting (see Schilling and Winchester, 1967, fig 2).

One important petrogenetic aspect of Rhum (and Hebridean) basalt trace element geochemistry, widely reported in the literature, is their depletion in incompatible elements (Ridley, 1973; Thompson *et al.*, 1980; Morrison *et al.*, 1980). Other alkalic basalt suites with similar "anomalously low" trace element abundances have been reported (e.g. Baxter, 1976). Morrison *et al.* (1980) proposed that the upper mantle beneath the Hebrides suffered depletion in these elements as the result of a previous melting episode. This may have involved a (<1%) melt fraction with extremely high incompatible element concentrations, representatives of such a liquid possibly being the Permian lamprophyre dykes. A similar model has been postulated for the Baffin basalts and picrites (O'Nions and Clarke, 1972) and the alkalic basalts of Mauritius (Baxter, 1976).

6.5.4 Implications for magmatic evolution

6.5.4.1 Composition of the parental liquid to the layered ultrabasic complex: The intrusion of high-MgO (picritic) liquids, albeit on a small scale, at a late stage in the evolution of the ultrabasic complex suggests either (i) such liquids were generated late in the development of the complex, e.g. at a culmination of heat production in the mantle melting event, or (ii) high-MgO liquids were generated throughout its history, only reaching upper-crustal levels prior to the development of magma reservoirs, or in the waning stages of magmatic activity when the chambers had largely solidified. Hot dense high-MgO (>9 wt%) liquid which entered a magma chamber containing lighter, fractionated basaltic (MgO <9 wt%) liquid, would have been "ponded" at the base of the chamber to form an independent layer which exchanged heat across a stable interface, but did not extensively mix with the resident magma (Huppert and Sparks, 1980a, b).

Obvious and very important petrogenetic implications of these two models concern the genetic relationship, if any, between the high-MgO liquids and the parental liquid to the L.U.C. Model (ii) implies that magnesian liquids may have been parental to part, if not all, of the ultrabasic sequence; model (i) suggests they were not.

The absence of chilled margins to the Rhum ultrabasic complex has given rise to considerable uncertainty and debate about the composition of the parent liquid. The proposed composition must take into account the following constraints, which this and previous studies (Brown, 1956; Dunham and Emeleus, 1967; Donaldson *et al.*, 1973; Donaldson, 1977a; Gibb, 1976; Henderson and Gijbels, 1976; Forster, 1978; Dunham and Wadsworth, 1978) place on the parent liquid. The liquid should (i) be capable of crystallizing in the sequence a) ol(+spinel) (b) plag and (c) cpx, with initial compositions Fo_{89}, Mg/Mg+Fe 0.4 and

Cr/Al 1.5, An_{92} and Ca_{46} Fe_6 Mg_{48} respectively (ii) be capable of producing the slight but significant cryptic variation described in Chapter 2, (iii) have a LREE-enriched chondrite-normalized pattern (Henderson and Gijbels, 1976), (iv) have a relatively high water-content and high normative percentage of olivine, to produce harrisitic textures (Donaldson, 1977a).

Proposed compositions are presented in Table 6.7. Brown (1956), on the basis of chemical and textural evidence, calculated that the parent liquid was a high-Al, tholeiitic basalt. The fine-grained olivine gabbro bears a close resemblance to the estimated composition.

Since this basaltic model was first put forward, several authors have suggested a more basic parent material. Drever and Johnston (1972), Donaldson *et al.* (1973), Donaldson (1975, 1977) and Gibb (1976) have proposed a highly olivine-phyric picrite or high-MgO basalt (which they refer to as "feldspathic peridotite" magma), and stressed what they regard as an important characteristic, the CaO-rich nature of the liquid ($CaO/CaO+Na_2O+K_2O > 0.8$). The latter was deemed necessary to explain the development of unzoned bytownitic spherulites (poikilo-macrospherulitic feldspars, Donaldson *et al.*, 1973) in the peridotitic igneous breccias, without the operation of an adcumulus growth process (since the breccia is envisaged as having been intruded as a crystal-mush and therefore precluding the possibility of diffusional exchange between intercumulus and supernatant liquids). However, in view of Irvine's (1980) model of infiltration metasomatism, adcumulus growth may indeed proceed deep within a crystal-pile, and consequently arguments for a low-alkali and high-CaO liquid may not hold. Meanwhile the discovery of a distinctive high-CaO (>12 wt%) low-alkali (<2 wt% Na_2O+K_2O) olivine tholeiite (Preshal Mhor) magma type on Skye (Esson *et al.*, 1975; Mattey *et al.*, 1977) led Donaldson (1977a) and Dunham

and Wadsworth (1978) to postulate a similar composition for the Rhum parent liquid. Such a liquid is also regarded as being parental to the Ben Buie layered gabbro, Mull (Skelhorn *et al.*, 1979) and the Slieve Gullion dolerites (Gamble, 1979).

Consideration of the constraints (i) to (iv) outlined above, narrows down the choice of plausible parental liquid compositions. With regard to composition of liquidus phases and their crystallization order, Brown (1956) and Gibb's (1976) hypotheses fulfill this constraint (i) , as confirmed experimentally at atmospheric pressure (Forster, 1978). In contrast, the melting relationships (1 atm) of two Preshal Mhor type basalts (Esson *et al.*, 1975) show plagioclase to be the liquidus phase, closely followed by olivine, then clinopyroxene and magnetite. The Preshal Mhor magma type (high-CaO, low-alkali tholeiite) does not therefore satisfy fundamental requirement (i).

With regard to the cryptic variation (constraint (ii)), a detailed discussion has been presented in Chapter 2 (section 2.10) with the bulk of the evidence favouring a high-MgO basalt parent (analysis 5, Table 6.7). In addition to its capability of crystallizing large quantities of olivine with only slight compositional variation (as a result of equilibrium crystallization), a picritic liquid also reduces the volume requirements of the parent liquid and the differentiates (expelled and removed by erosion prior to eruption of the Rhum lavas, section 6.5.4.2).

With respect to constraint (iii), the L.U.C. parental liquid is highly likely to have been LREE-enriched in view of the LREE-enrichment of the cumulus plagioclase (Henderson and Gijbels, 1976). The Preshal Mhor magma type, with a LREE-depleted pattern, does not satisfy this requirement.

If criterion (iv) - high MgO-(>20% normative olivine) and water-contents - is indeed necessary for the formation of widespread harrisite layers in the L.U.C. (Donaldson, 1975a), the Preshal Mhor basalt composition must be excluded on the grounds of being insufficiently magnesian, having less than 15% normative olivine.

Since the Preshal Mhor-type magma has not been recognized within the Rhum dyke swarm (the small number of Preshal Mhor type dykes cutting the late-stage lavas are regarded as outliers of the main Skye swarm), it may not have been available during formation of the L.U.C., and this widely favoured contender for parent liquid, in addition, fails to satisfy the four requisite criteria.

In conclusion, the existence of high-MgO liquids (9-20%) on Rhum has been demonstrated (section 6.5.1) and it is suggested that the primary magmas generated beneath the complex involved liquids with >20% MgO.

6.5.4.2 Relationship to the lavas of Rhum and Skye: A possible genetic relationship between the Rhum lavas (Emeleus, 1976; Dunham *et al.*, in press) and the L.U.C. was discussed by Dunham and Emeleus (1967). Assuming Brown's (1956) proposed high-Al basaltic parental composition to be correct, the authors accounted for the absence of tholeiitic lavas supposedly erupted during formation of the layered intrusive complex, by extensive erosion prior to eruption of the present lavas. The possibility that the Rhum mugearites are residual material from the magma chamber was briefly mentioned, but rejected on the grounds that the Rhum magma would have to be alkali basalt with a high proportion of Mg-rich olivine, and the ultrabasic cumulates made up of peridotite and allivalite in approximately equal amounts.

Ridley (1973) likewise placed great significance on the relative abundance of mugearite in the lava pile, but argued that their close

association with alkali olivine basalt suggested a genetic connection between them. He noted that the alkali olivine basalts are chemically similar to the high-Al parental basalt of Brown (1956), and therefore concluded that the observed lavas appear to be chemically related to the layered series.

Forster (1980), using unpublished data of Dunham *et al.* (in press) outlined several geochemical differences between the dykes and lavas (the latter have less Mg, Ca, Ni and Cr) and suggested the dykes fed, not the lavas now preserved in situ, or in the interbedded conglomerates (see Chapter 1), but earlier lavas subsequently eroded. Detailed investigations of the lavas and contents of the Tertiary conglomerates are underway (Dunham *et al.*, in press). The conglomerates are a valuable key to a better understanding of the geological history of Rhum.

Several lines of evidence suggest that the Skye igneous centre is younger than Rhum.

(a) the existence of Skye Main Lava Series and Preshal Mhor-type dykes cutting the Rhum lavas, implies that part, if not all, of the Skye Main Lava Series post-dates the Rhum complex. Evidence concerning the relative ages of the Skye and Rhum lavas is sparse and equivocal - see (b) and (c) below.

(b) K-Ar dates for unaltered Skye lavas (59.2 ±1 m.y., Taylor and Forrester, 1971) and the Rhum Western Granophyre (58 m.y., Emeleus pers. comm.) would suggest that the two are contemporaneous, and therefore the Skye lavas pre-date those of Rhum. However:-

(c) granophyre pebbles within the Skye Tertiary conglomerates are most probably derived from Rhum (see Chapter 1), and therefore imply that Skye lavas are younger than the Rhum central complex. The Rhum lavas may be broadly contemporaneous with the Canna and Skye sequences, bearing in mind that there are several erosional intervals of unknown magnitude in the remaining Rhum lava series.

In conclusion, the bulk of the available evidence suggests that the Rhum granophyres, L.U.C. (and dykes) were uplifted and eroded prior to eruption of the Skye (and ?Rhum) lava piles, and hence that mantle fusion in the Rhum area pre-dated that of Skye.

6.5.4.3 The Tertiary igneous cycle in Rhum: comparison with that of Skye: The history of igneous events in Rhum (Dunham and Emeleus, 1967; Forster and Emeleus, 1979) may be simplified to the following sequence: acid magmas of the high-level granophyres and felsites — picritic magmas of the central complex — basic/picritic dykes — lava series — Preshal Mhor-type dykes cutting the lavas. It contrasts markedly with the sequence in Skye (Thompson *et al.*, 1980): Skye Main Lava Series — Preshal Mhor-type basalts — basic magmas of the Cuillin pluton — acid magmas of the high-level granites — basic dykes cutting the granites. As discussed in section 6.5.4.2, the last two events in the Rhum sequence are possibly contemporaneous with the initial two in Skye.

From geological evidence, the formation of the L.U.C. (quasi-contemporaneous with the granophyres) appears to have been the first major Tertiary basaltic magmatic event in Rhum. Estimated MgO-contents of the liquids in equilibrium with the most Fo-rich olivines in the E.L.S., W.L.S. and C.S. are shown in fig 6.21. These estimates have been made using data from phenocryst - matrix pairs in rapidly-cooled dykes. Since the cumulus olivines may have crystallized under equilibrium conditions (Chapter 2, section 2.10), and suffered slight sub-solidus re-equilibration involving a decrease in Fo-content (Roeder *et al.*, 1979), the MgO estimates (11.5-15.0 wt%) for the L.U.C. liquids are too low and can only be taken as minimum values. As can be seen from fig 6.21 there is a progressive increase in the relative MgO-contents of successive liquids supplying high crustal levels, reaching

a maximum of ca. 20.5% in the dyke suite. This trend can be interpreted in one of two ways.

(a) it implies "top to bottom" emptying of a "stratified" magma reservoir (dense picritic liquid underlying less dense basaltic liquid). Such a chamber may have been situated in the lower crust.

(b) it suggests a progressive increase in the degree of partial melting.

The parallelism of the LREE patterns for the plagioclase from successive units in the E.L.S. (Henderson and Gijbels, 1976) suggests a relatively homogeneous source region, and precludes (b). Clearly REE data for feldspar from the W.L.S. and C.S. are needed to further evaluate the possible role of varying fusion. Ultimately the solution may involve a combination of (a) and (b). Forster (1980) reports a concomitant increase in alkalinity of the dykes with decreasing age, which suggests a general increase in the depth of melting (limited melting at greater depths giving more alkalic liquids). Alternatively, these dykes may represent derivatives of the picritic primary liquid, which stagnated in the upper mantle and fractionated to produce *ne*-normative, high-alkali liquids.

The extrusion of transitional LREE-enriched Skye Main Lava Series basalts appears to have been the first Tertiary igneous event in Skye. Primary liquids were formed during small degrees of melting of garnet lherzolite at a depth of ca. 65 km (Thompson *et al.*, 1972; Thompson, 1974). The indistinguishable $^{87}Sr/^{86}Sr$ ratios and Th/Ta and Ta/Hf ratios of the Preshal Mhor basalt-type (occurring as several flows in the stratigraphically-higher parts of the Skye lava pile (Esson *et al.*, 1975) and dykes which are believed to have fed upper, now eroded, portions of the pile (Mattey *et al.*, 1977)) imply that these two main basalt types were produced from the same volume of upper mantle

(Thompson *et al.*, 1980). The Preshal Mhor magma type is regarded as the product of a subsequent further increment of fusion of the lherzolite, in which all the Ca-rich clinopyroxene was consumed (Thompson *et al.*, 1980). Clearly the Skye melting event proceeded a stage further than that of Rhum, to the generation of further MgO-rich (picritic) liquids, which separated olivine during ascent, to erupt as Preshal Mhor-type tholeiites. Several authors (Esson *et al.*, 1975; Mattey *et al.*, 1977) contend that the Preshal Mhor liquids may have been parental to the basic-ultrabasic Cuillin intrusive complex.

6.5.4.4 An analogue to mid-ocean ridge basalt genesis: The close chemical resemblances (e.g. major and trace element abundances and ratios) between the picritic-basaltic magmas of Rhum and the high-MgO basalts from known Tertiary spreading centres, e.g. Baffin Bay, raises the question of whether there were genuine (but abortive) attempts to produce oceanic crust beneath Rhum. Such a process is concluded to have occurred (to a slightly more advanced stage) beneath Skye: the high-CaO, low-alkali olivine tholeiites (Preshal Mhor type) strongly resemble mid-ocean ridge basalts from Iceland and the Mid-Atlantic Ridge (in major element chemistry (Esson *et al.*, 1975), and in low incompatible element abundances and LREE-depleted patterns (Mattey *et al.*, 1977)). Magmas of this character were not uncommon in the British Tertiary Volcanic Province (Drever and Johnston, 1966; Thompson *et al.*, 1972; Esson *et al.*, 1975; Mattey *et al.*, 1977; Donaldson, 1977; Gamble, 1979) and possibly represented the final vestiges of an early Tertiary thermal event when an unsuccessful attempt at crustal rifting took place in NW Britain. The incipient rift zone may be outlined by the NNW-SSE alignment of the central volcanic complexes.

6.6 Conclusions

1. The Rhum minor intrusion suite is composed of transitional basalts (<9 wt% MgO) and picrites (>9% MgO). Both groups contain mildly *ne*- and *hy*-normative members.
2. MgO-contents of the liquids vary from <6% to 20.5%.
3. Major element variations are controlled by the fractionation of olivine, Al-rich spinel (at high pressures only), Fe-Cr spinel (at low pressures only), plagioclase and minor clinopyroxene - in that order.
4. Major element compositions of the more fractionated dykes are comparable to the transitional basalts of Skye and Mull, but have the higher CaO- and lower alkali-contents (>12% and <2% respectively) more typical of the Preshal Mhor magma type.
5. Trace element chemistries are also closely comparable to other Hebridean transitional basalts: contents of incompatible elements are very low, and chondrite-normalized REE patterns are slightly LREE-enriched.
6. No dykes with the distinctive trace element chemistry (e.g. high Y/Zr ratios, 0.5, and LREE-depleted patterns) of the Preshal Mhor magma type have been found within the L.U.C.
7. The weight of evidence (petrographic, mineralogical and geochemical) is in favour of a highly magnesian (ca. 20.5% MgO) parental liquid for the dyke suite.
8. The high-MgO liquids are believed to be the result of small degrees of melting (15-20%) of garnet lherzolite at a depth of ca. 65 km (20 kb), leaving a lherzolite residuum.
9. Similar magnesian liquids were parental to the layered ultrabasic complex.
10. The Rhum Tertiary melting event pre-dates that in Skye.

CHAPTER 7

STRUCTURAL AND PETROLOGICAL EVOLUTION OF THE RHUM ULTRABASIC COMPLEX

The following sections present:

1. A summary of the field, petrographic and chemical data pertinent to the stratigraphic relations within and between the various layered series in the complex.
2. A two-stage model for the igneous history of the complex based on the above data, and additional data on associated minor intrusives (gabbros, dykes and peridotites: Chapters 3-6).
3. A tectonic model compatible with rifting processes, which can account for the emplacement and present distribution of the layered series within the complex.

7.1 Stratigraphy and Petrologic Relationships

In the ultrabasic rocks of the Barkeval-An Dornabac area three major sequences have been distinguished on the basis of field mapping (fig 2.1): (i) the westerly continuation of the E.L.S. (Units 4-12 only), (ii) the northern extension of the Ard Mheall Member of the W.L.S. (see section 2.5), and (iii) the newly-defined C.S. (see section 2.6.1).

Despite the textural and mineralogical affinities of the W.L.S. with the E.L.S., several significant differences exist between the two cumulate sequences (summarized in section 2.2.1), which must be explained by any proposed model. The differing thicknesses of cyclic units may be related to the volume of the magma influxes and the time interval before the next magma replenishment (larger and longer respectively, in the W.L.S.). The absence of thick plagioclase cumulates, and the remarkable development of harrisite (often comb-layered) in

the W.L.S. probably reflect, in addition to possible compositional differences, fundamental differences in the crystallization conditions (e.g. the state of convection; rate of heat loss through walls, floor and roof; efficiency of volatile concentration).

Contact relations indicate that the W.L.S. and E.L.S. were intruded by the Central Series, and therefore (a) suggest a two-part history for the complex, and (b) raise the question of how many magma types were involved in its evolution.

Closely comparable mineral composition data for the E.L.S. and C.S. (sections 2.9 and 2.10) imply that the two series were derived from a similar magma type. From the available published data (Wadsworth, 1961) there is no necessity to invoke a different parental magma composition for the W.L.S.. Therefore during the formation of the three layered series the composition of the magma entering the Rhum depositional chamber appears to have varied very little with time (Chapter 6, fig 6.21).

The apparent lack of significant variation in the parental magma composition for the E.L.S. and W.L.S., and the possible explanation of the observed petrological differences in terms of physio-chemical conditions in the chamber, suggest that the two sequences represent uplifted portions (along the Inner Ring Fault, fig 2.1) of the original intrusion (as suggested by Wadsworth, 1961). The continuity of strike of the igneous layering between the two series, such that they appear to define a single W-E elongated body (fig 7.1A), would suggest (a) that the original intrusion may have had much the same dimensions as the now-exposed portion (i.e. 7 km diameter), and (b) that the amount of relative vertical displacement of the two parts may have been minimal. If (b) is correct the differences between the E.L.S. and W.L.S. would require explanation in terms of lateral variation in

crystallization conditions etc. over distances of 2-3 km (the width of the intervening Central Series). This may not be plausible.

The nature of the magma entering the Rhum chamber varied greatly. Fo-NiO data for the E.L.S. olivines, and the evidence of in situ fractionation from cryptic variation patterns (Chapter 2, and Dunham and Wadsworth, 1978) imply that the magma supplying the chamber carried only minor amounts of liquidus olivine (and spinel) - ca. 10%. A similar conclusion probably holds for the W.L.S.. In contrast, several lines of evidence (field, textural and chemical - sections 2.6, 2.7.3.9 and 2.9.1 respectively) imply the magma entering during formation of the C.S. was highly olivine-phyric (<ca. 50% crystals).

7.2 Stratigraphic and Petrological Model

With the above in mind, together with the conclusions presented in each chapter, the following generalized history is proposed for the Rhum ultrabasic complex.

It is proposed that the parental liquid to the complex was of a transitional picritic (ca. 20.5 wt% MgO) composition, and the result of small degrees of melting (15-20%) of garnet lherzolite at a depth of ca. 65 km (20 kb) leaving a lherzolite residuum. This liquid was forced into a feeder system to a developing intrusive chamber at, or near, the base of the Hebridean crust (25-30 km depth). Due to the density-filter effect imposed by the crust, the high-MgO liquid was ponded at its base (ascent and eruption were limited). Within this holding chamber the parental liquid underwent fractionation of predominantly olivine (and Fe-Ni sulphides) to form ultramafic cumulates (e.g. dunites with ca. Fo_{92}-Fo_{90}).

STAGE 1: At the same time a second upper-crustal magma chamber was largely established, and for an extended period was more or less

"open" to appreciable additions of magma ascending from the lower chamber (ascent of the magnesian liquid was possible once fractionation had lowered the density sufficiently). The magma supplying the upper chamber had a minimum MgO-content of ca. 15% (estimated from data from phenocryst-groundmass pairs in rapidly-cooled dykes, using Fo_{89} the most magnesian olivine composition recorded in the E.L.S. (and W.L.S.), and conditions during ascent were such that it carried only a small proportion of olivine crystals (ca. 10%).

Multiple replenishment, magma mixing, in situ fractionation and crystal deposition within this high-level intrusion produced a thick sequence of olivine, plag-ol±cpx cumulates. The residual low-temperature magma (basalt, <9 wt% MgO) was extruded intermittently during contemporaneous surface volcanic activity. Occasional batches of the basaltic magma intruded the consolidated cumulate pile (gabbroic minor intrusions, Groups 2 and 3). Eventually magmatic activity waned and the layered intrusion cooled and solidified. Portions of the original intrusion were tectonically emplaced (see sections 7.3 and 7.4) - now exposed as the Western and Eastern Layered Series.

STAGE 2: After an unknown (but probably relatively short) time interval magmatic activity in upper and lower crustal levels resumed (a continuance of the mantle thermal event in stage 1, but marked by slightly increased depth and degree of melting). As in stage 1 transitional high-MgO liquid fed into the lower magma reservoir. However, instead of ponding and fractionating, this liquid (with ca. 20% MgO) ascended readily and rapidly to upper crustal levels, aided by strong crustal fracturing and the absence of intervening reservoirs (potential magma "traps"). As a result of (a) olivine crystallization, (b) a slow rate of cooling, and (c) limited net loss of crystals, during the ascent along a probably pre-heated feeder system, the magma

reached upper crustal levels carrying up to 50% olivine crystals (petrographic evidence from the picritic dykes supports the deliverance of highly olivine-phyric magma to the upper crust environment).

Successive batches of the porphyritic magma were emplaced into the cumulates (E.L.S. and W.L.S.) of the earlier intrusion (stage 1), and underwent crystal accumulation and in situ fractionation to form the Central Series cumulates.

Initial batch(es) brecciated the E.L.S. and W.L.S., forming the Outer Breccia Member (see An Dornabac - Barkeval cross-section, in pocket at back of Volume 2). Subsequent influxes of magma spread laterally over the breccia and possibly onto the W.L.S. and E.L.S. to produce the Dornabac and Long Loch Members (fig 7.1B, and Section 1 in pocket). Eventually the feeder system "choked", and when magmatic activity resumed material was emplaced into the earlier cumulates - hence the transgressive nature of the Ruinsival Member (fig 7.1C).

Intrusive offshoots from the Central Series porphyritic (picritic) magma are represented by the numerous non-layered peridotitic minor intrusions, and low-temperature (basaltic) magma by the gabbroic minor intrusions, Groups 4-6.

7.3 Structural Relationships

The Eastern and Western Layered Series ultrabasic rocks together define an E-W elongated intrusion (see fig 7.1A), and marginal relationships imply that it was emplaced in an essentially solid condition along a ring fracture (Inner Ring Fault). Basaltic magma which, according to Brown (1956) lubricated the mass during emplacement, solidified to form the Marginal Gabbro. The amount of uplift is indeterminate but was probably quite small (section 1.3.2).

Detailed mapping of the An Dornabac-Barkeval area has revealed

the significant role played by the Long Loch Fault in the development of the Central Series, and the relevance of the Series to the evolution of the complex.

At its northern and southern extremeties the Central Series cuts the Inner Ring Fault (figs 2.1 and 7.1) and therefore was formed after the tectonic emplacement of the E.L.S. and W.L.S.. The latter two Series are nowhere found in contact and their structural relationship remains unresolved.

7.4 Structural Model

A model is presented for the geometry of the three layered series. In simplest terms the Rhum ultrabasic complex consists of two cross-cutting intrusions: the elliptical (E-W elongated) body defined by the E.L.S. and W.L.S. (Stage 1 of the petrological model) is disrupted and cut by the N-S elongated dyke-like mass of the C.S. (Stage 2 of the petrological model) - as shown in fig 2.1 and Section 1 (in back pocket).

The increase in dip of both the E.L.S. and W.L.S. igneous layering towards the contacts with the C.S. (e.g. on the southern face of Barkeval, and in the Ard Mheall Member, Map 1 and Section 1) may or may not be related to the formation of the C.S.. Differential compaction within the Stage 1 intrusion may have produced this configuration prior to the onset of C.S. formation. Alternatively the case can be envisaged where the interior of the uplifted E.L.S.-W.L.S. mass was still relatively hot and semi-consolidated, and therefore capable of deforming during the Stage 2 (C.S.) phase of intrusive activity.

The following points suggest an important role was played by the Long Loch Fault.

(a) field relationships indicate that the C.S. has a dyke-like form

and is asymmetrically developed about the fault (Section 1).

(b) the geographical distribution of the Series is closely-linked to the fault (fig 2.1 and 7.1).

It is therefore proposed that during Tertiary times the Long Loch Fault was a major line of crustal weakness, extending deep into the Hebridean crust, and capable of tapping magmas from lower crustal holding chambers (see section 7.2) or directly from the upper mantle.

The maximum development of the C.S. cumulates (520 m stratigraphic thickness), together with the large positive gravity anomaly (McQuillan and Tuson, 1963), at the southern end of the Long Loch Fault suggests that the main conduit was sited in this region. Some magma moved laterally northwards along the fracture, hence the apparent overlapping of the Long Loch Member onto Harris Bay (W.L.S.), Outer Breccia and Dornabac Members, and the rapid thinning of the latter two northwards into a thin terminating wedge - the Minishal tongue (fig 2.1 and Map 1).

Contemporaneous disturbance and subsidence in the axial region of the "dyke" during accumulation of the Dornabac and Long Loch Members produced the steeply-dipping igneous layering in these rocks (sections 2.6.3.2 and 2.6.4).

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APPENDIX A

ELECTRON MICROPROBE ANALYSIS

All mineral analyses presented in this thesis were obtained using a Cambridge Instruments Microscan 5 electron microprobe at the Grant Institute of Geology, Edinburgh. The two methods of X-ray analysis available, wavelength-dispersive and energy-dispersive, have been used. Details of each are given below.

Following petrological examination of the polished thin sections, a conductive, vacuum-deposited, carbon coating was added. Standards and sections were coated simultaneously.

A.1 Wavelength-Dispersive Analysis

Silicate and oxide analyses were obtained by wavelength-dispersive spectrometry (WDS) using a gun potential of 20kV, a beam current of 30 nA and a beam diameter of ca. 1.5 μm. Since a maximum of four elements may be determined in one group, accurate centring and relocation of the electron beam is essential for successive element groups. Although more time-consuming than the energy-dispersive system (EDS), this method allows for the variation of counting times, as required by the analytical precision. Precision given by WDS is better for minor and trace elements.

In general, background counts (either side of the peak) were only measured on the standards and on one example of each phase in each polished section, unless a phase was shown to have a wide compositional variation. Analytical conditions are summarized in Table A.1.

The calculation of apparent concentrations (relative to the average of standard counts and including a correction for "deadtime") was carried out using the APPCONC computer program written by D. J. Humphries. In general, only two sets of standard count rates (before and after each set of unknowns) were used, though in rare cases,

e.g. if a significant drift in standard count rates was found, the standards were read more frequently.

Further data reduction and correction involved use of the PROBE computer program (written by D. J. Humphries): the atomic number, absorption, and fluorescence (ZAF) corrections follow the proceedures of Sweatman and Long (1969). Precision and detection limits for the various phases analysed, are shown in Table A.2.

A.2 Energy-Dispersive Analysis

Sulphide minerals were analysed by energy-dispersive methods, using a Si(Li) Link Systems detector fitted to the Microscan. The gun potential remained unchanged at 20kV, but a smaller beam current (6 nA) and beam diameter (1.0 μm) were used than for WDS. Live-times were 100 seconds, and the processing of collected spectra was carried out on-line by a Data General Nova Computer, using a program written by Statham (1975), and the ZAF correction proceedures of Sweatman and Long (1969).

Short-term drift was measured and corrected for, by reference to a Co-metal monitor. The EDS only needed to be fully calibrated at infrequent intervals. The sulphide phases were analysed by EDS since good precision and detection limits of minor and trace elements were not required.

A.3 Sampling

All samples were collected by the author from surface exposures. In accordance with the aims of the Nature Conservancy Council's Chief Warden, regarding the conservation of sites of geological interest, hammering of surface exposures was kept to a minimum. A map showing "prime" sites of geological interest was compiled, and areas graded according to their vulnerability to hammering, photogenic appeal etc. As a direct result of this exercise, several geological localities

have been designated "protected sites", and hammering forbidden (except for collection of research/teaching samples).

A.4 Key to Analysis Tables in Appendix A.

See Table A.3 for explanation of code letters, and lists of elements analysed.

APPENDIX B

X-RAY FLUORESCENCE ANALYSIS

B.1 Preparation of Whole Rock Powders

All samples collected in 1979 by the writer, were prepared for analysis at the Grant Institute, Edinburgh. To obtain representative analyses of coarse-grained ultrabasic and gabbroic rocks, samples of at least 500 g were collected where possible. 200 g samples of the homogeneous, fine-grained dykes were collected.

Using a diamond saw, the samples were trimmed to remove all weathered surfaces, cut into 1 cm-thick slabs, then rinsed in clean running water, and finally split in a Cutrock Engineering hydraulic splitter. The rock fragments were then crushed in a Sturtevant jaw-crusher, to 0.5 mm chips and powder. Samples were then ground in a Tema tungsten-carbide swing-mill to <100 mesh. Some oxidation of ferrous iron in the samples may occur during this mechanical grinding (Fitton and Gill, 1970) and the Tema mill contaminates samples with cobalt and tungsten. Neither element was analysed in this study.

B.2 Analytical Procedure - Preparation

All elements were determined using the Philips PW 1450/20 sequential automatic X-ray fluorescence spectrometer at the Grant Institute of Geology, Edinburgh. Only a summary of the techniques is given here. For full details see Thirlwall (1979).

B.2.1 Major elements

Major element analyses were carried out on fused glass discs prepared by a method similar to that of Norrish and Hutton (1969). The essentially homogeneous nature and reproducible surface finish of these discs are necessary to minimize the effect of sample surface inhomogeneities, on the weakly penetrative long wavelength radiation used.

Small glass jars of sample powders were dried overnight in an oven at 110°C to remove adsorbed water (H_2O^-). Samples were not ignited prior to initial flux addition (potential weight losses on ignition - LOI - were <5%). Spectroflux 105, a lithium tetraborate-lithium oxide-lanthanum oxide mixture was added to 1.05 g of sample powder, in a fixed sample: flux ratio (1:5.333) and the mixture fused at 1100°C until completely dissolved (approximately 20 minutes).

When cooled to a glass, the sample (plus crucible) was reweighed, and a small amount of flux (ca. 0.01 g) added to make up for flux volatile loss. The crucible was then transferred to Meker burners (at approximately 1000°C) to re-fuse the extra flux. After swirling to ensure homogeneity, the melt was cast into a glass disc, using graphite plattens and stainless steel rings on a 240°C hotplate. The quenched discs were covered, allowed to cool, and all glass chips kept with each disc.

B.2.2 Trace elements

Trace element analyses were carried out on undiluted pressed powder discs, with no problems regarding heterogeneity or quality of the surface finish. Approximately 7 g of sample (calculated by Fitton, pers. comm. 1979, to allow 99% NbK radiation in a matrix with very low absorption) were mixed with 6 drops of a binding agent (2% aqueous solution of polyvinyl alcohol) and transferred to polished tungsten-carbide pressing apparatus, and hand-pressed. Backing to the pressed powder pellet was provided by powdered boric acid, and the complete pellet compressed to about 10 ton/in^2 for ca. 2 minutes, using a hydraulic press. The apparatus was thoroughly cleaned, with alcohol, between samples.

B.2.3 The Philips PW 1450/20 spectrometer

Except for the manual selection of the high voltage X-ray tube, the PW 1450/20 spectrometer is automated, and runs on three programs.

Program 90: measures counts for major elements (Na, Mg, Al, Si, P, K, Ca, Ti, Mn, Fe) using a Cr-anode X-ray tube in an evacuated X-ray path.

Program 91 - Cr tube: measures counts for the trace elements Sc, V, Cu, Ba and La.

Program 92 - W tube: measures counts for the elements Ni, Zn, Th, Cr, Rb, Sr, Y, Zr, Nb, Ce, Sm, Nd using a tungsten-anode X-ray tube in an evacuated X-ray path.

Analytical conditions are summarized in Table B.1; the various interferences between analytical lines, and their treatment, are given in Table B.2.

B.3 Processing The Data

B.3.1 Major elements

Major element calibrations were based on international standards. No corrections for mass absorption differences were necessary because the samples have similar mass absorption coefficients to the standards, (as a result of the heavy absorber La in the flux). Averaging of count ratios, and calibration regressions were carried out using the computer programs RATMAJ and MAJORS respectively (written by Thirlwall, 1979).

B.3.2. Trace elements

Trace element calibrations were based on international standards and spiked synthetic standards (Thirlwall, 1979). Mass absorption coefficients were calculated for each sample, on the basis of the major element analyses. Averaging of count ratios, calibration regressions, and interference and matrix corrections were carried out using

the computer programs CRTRACE, WTRACE and TRACE respectively (written by Thirlwall, 1979).

B.4 Precision And Accuracy

The reproducibility of fused glass discs and pressed-powder pellets has been investigated by Thirlwall (1979), by the preparation and analysis of six discs and six pellets from one sample. The results are given in Table B.3. Table B.4 presents data on the analytical precision of the methods, collected by the repeated analysis of one disc and pellet (Thirlwall, 1979). Accuracy (Table B.5) has been estimated by analysis of standard samples (Thirlwall, 1979).